

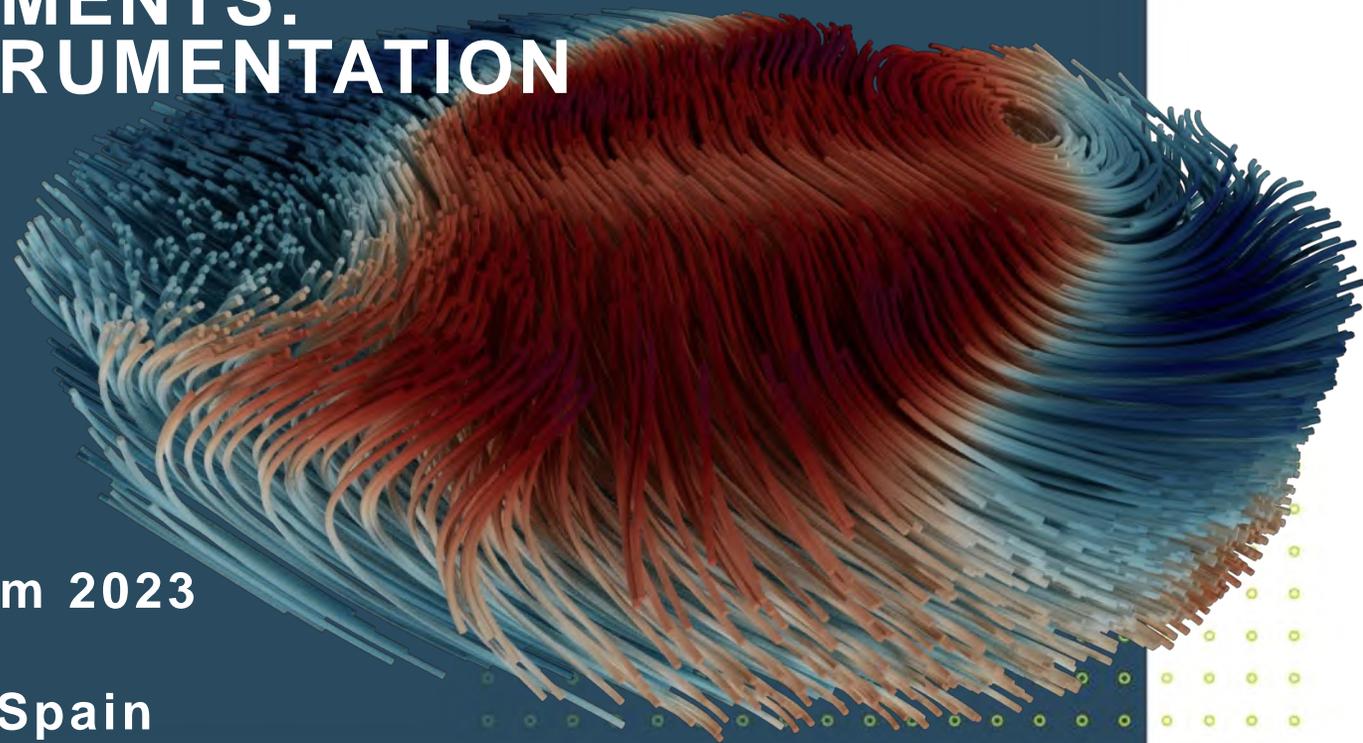


MAGNETIC MEASUREMENTS: ADVANCED INSTRUMENTATION

Claire Donnelly

European School of Magnetism 2023

7th September 2023, Madrid, Spain





OUR QUESTIONS FOR THIS MORNING'S LECTURES:

What do we need to consider?

Spatial resolution ✓

Sample environments

Time resolution

What methods are available?



Choosing the method for me and my samples?



	MFM	Nitrogen vacancy	TEM	MOKE	XMCD (synchrotron)	XMLD (synchrotron)	Spin pol. STM	SEMPA
Contrast	#, surface charges	#, surface charges	β , \perp , k	m	m, β , k	Niell vector (m, \perp , k)	m	m
Spatial resolution	10s of nm	10s of nm	Single digit nm (or below!)	100s nm - μ m	10s nm (& below!)	10s nm (& below!)	0.1 nm	~5 nm
Depth sensitivity	Surface sensitive	Surface sensitive	Thin samples ~100 nm	Surface sensitive	nm - μ m	nm - μ m	0.1 nm	1 nm
Sample environment	Field, cryo, electrical contacts	Field, -cryo, electrical contacts (in dev.)	-cryo, electrical contacts (in dev.), Fields challenging	Field, -cryo, electrical contacts (in dev.), TR	Field, -cryo, electrical contacts (in dev.), TR	Field, -cryo, electrical contacts (in dev.), TR	Cryo, field	Challenging, UHV & preparation required
Invasive	Yes	No	No	No	No	No	No	No
Sensitivity	Medium	High!	Medium	High	High, element sensitive	Medium, element sensitive, antiferromagnets!	High	High
Cost/ accessibility	Lab-based, accessible	Lab-based, recent commercial examples	Lab based, specialised equipment (10 ⁷ €)	Lab based, accessible	Large scale user facility, Open to all	Large scale user facility, Open to all	Lab-based, Specialised UHV equipment	Lab-based, Specialised UHV equipment



OVERVIEW OF (A SELECTION OF) AVAILABLE METHODS

	MFM	Nitrogen vacancy	TEM	MOKE	XMCD (synchrotron)	XMLD (synchrotron)	Spin pol. STM	SEMPA
Contrast	H , surface charges	H , surface charges	$B \perp k$	m	$m \parallel k$	Néel vector ($m \perp k$)	m	m
Spatial resolution	10s of nm	10s of nm	Single digit nm	100s nm – μ ms	10s nm (&	10s nm (& below!)	0.1 nm	~5 nm
Depth sensitivity								nm
Sample environment								challenging, HV & preparation required
Invasiveness								
Sensitivity					sensitive	sensitive, antiferromagnets!		gh
Cost/accessibility	Lab-based, accessible	Lab-based, recent commercial examples	Lab based, specialised equipment (10 ⁶ €)	Lab based, accessible	Large scale user facility, Open to all	Large scale user facility, Open to all	Lab-based, Specialised UHV equipment	Lab-based, Specialised UHV equipment

Beyond static imaging?



First video “The Horse in Motion” by Eadweard Muybridge, 1878





OUR QUESTIONS FOR TODAY:

ADVANCED INSTRUMENTATION? → BEYOND 2D IMAGING...

Higher dimensional investigations?

Vectorial imaging

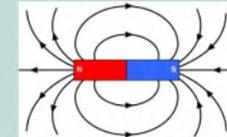
Magnetisation dynamics

Advanced sample environments

Temperature



Magnetic field



Current

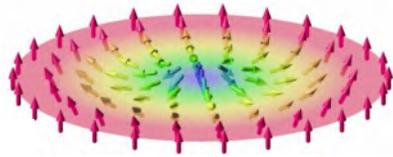


Pressure/ strain



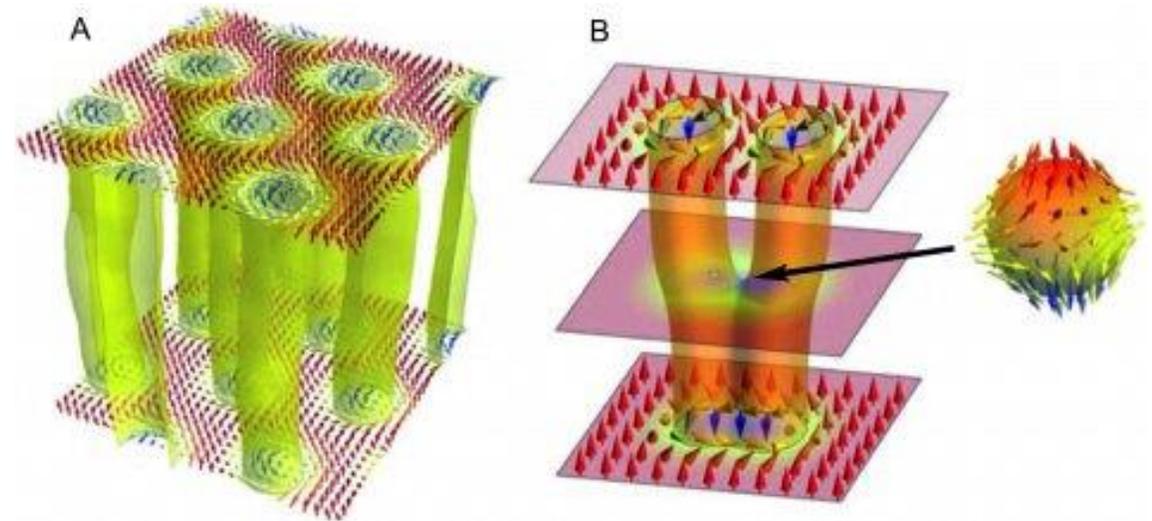
FIRST: WHY DO WE NEED VECTORICAL IMAGING? → THREE DIMENSIONAL TOPOLOGICAL TEXTURES

Skyrmions:

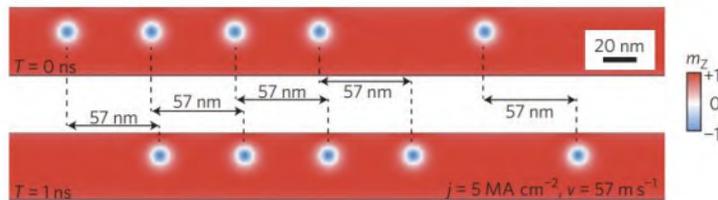


P. Milde et al., Science 340, 6136, (2013).

Well established
 Typically found in chiral systems



P. Milde et al., Science 340, 6136, (2013).



Fert et al., Nature Nanotechnology 8, 154 (2013)

Proposed racetrack devices

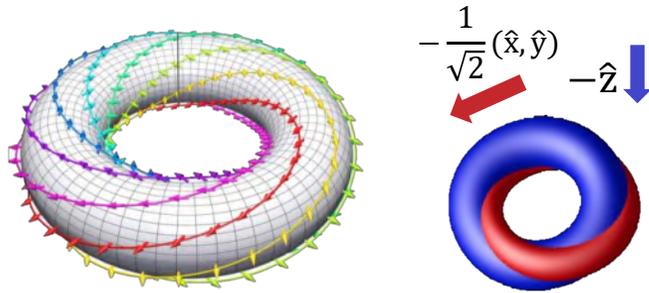
In 3D:

Prospect for complex 3D structure
 & topological transformations

FIRST: WHY DO WE NEED VECTORICAL IMAGING? → THREE DIMENSIONAL TOPOLOGICAL TEXTURES

Hopfions:

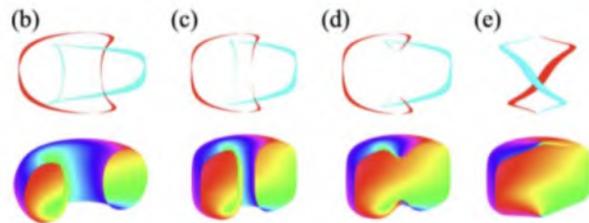
Growing number of theoretical works:



Rybakov et al.,
arXiv:1904.00250

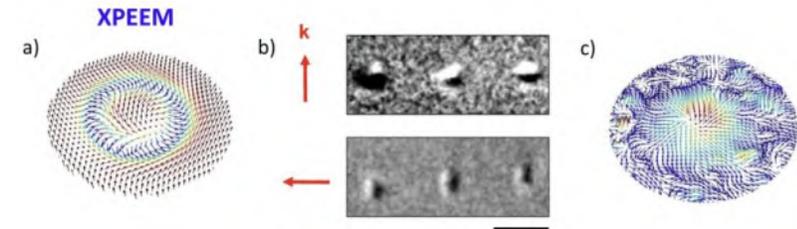
Sutcliffe PRL 118,
247203 (2017)

Statics, & dynamics:

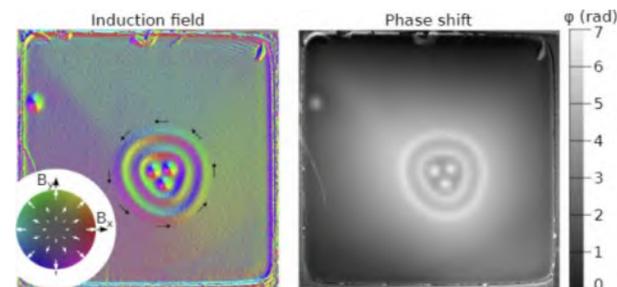


Liu et al., PRB 98 174437 (2018)

First experimental observations

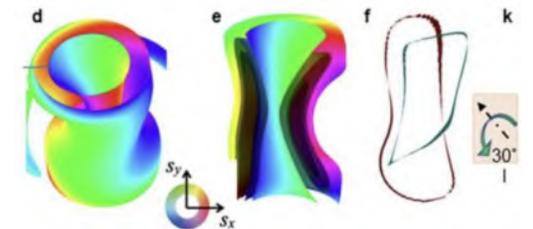


Kent et al., Nat Comm. 12 1562 (2021)



Kiselev et al.,
<https://doi.org/10.21203/rs.3.rs-2681064/v1>

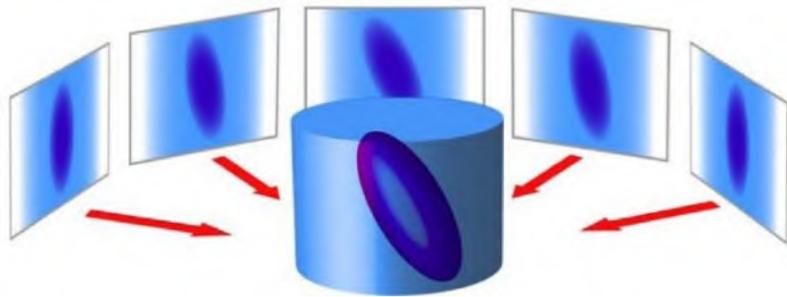
Fractional hopfions:



Yu et al., Adv. Mat. (2023)

3D MAGNETIC IMAGING

3D imaging: computed tomography



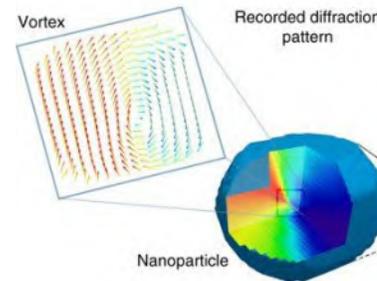
Guizar-Sicairos et al. Optics Express 19, 21345 (2011)

Recover 1 scalar value for each pixel
e.g. electron density

Tensor/ Vector tomography:

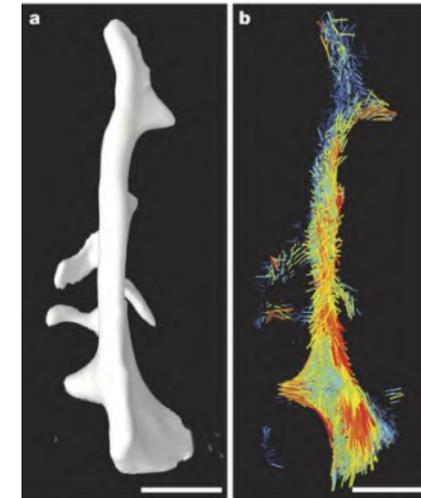
Multiple components for each pixel.

- Need more data – but what?
- Need new, tailored algorithms



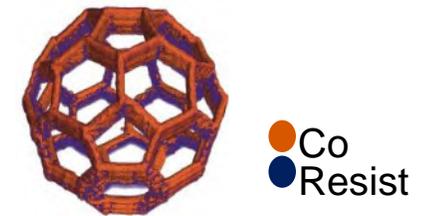
Imaging ferroelectric domains

Karpov et al., Nat.Comm. (2017)



3D SAXS tensor tomo

Liebi et al., Nature (2015)

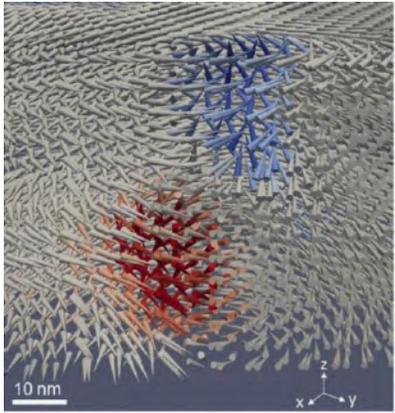


Element-specific tomo

Donnelly et al., PRL (2015)

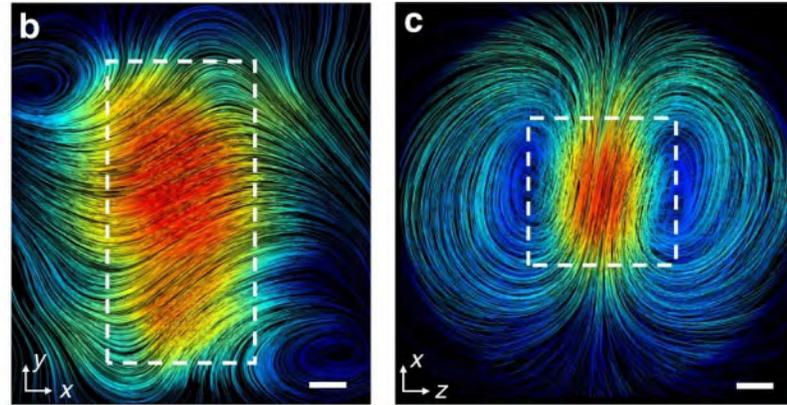
MAGNETIC IMAGING IN 3D

With electrons:



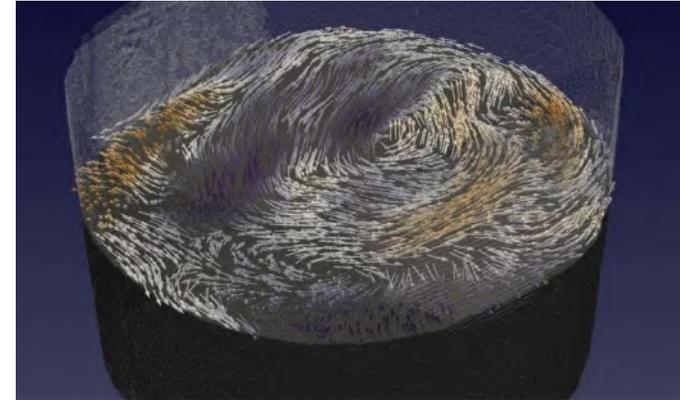
Tanigaki et al., Nano. Lett. **15**, 1309 (2015)
 Wolf et al. Chem. Mater. **27**, 6771 (2015)
 Wolf et al., Comm. Phys. **2**, 87 (2019)

... neutrons:



Hilger et al., Nat. Comm. **9**, 4023 (2018)
 Manke et al., Nat. Comm. **1**, 125 (2010)
 Kardjilov et al., Nat. Phys. **4**, 399 (2008)

... & X-rays:



Streubel et al., Nat. Comm. (2015)
 Blanco-Roldan et al, Nat Comm. (2015)
 Donnelly et al., Nature (2017)
 Suzuki et al., Appl.Phys. Expr. (2018)
 Hierro-Rodriguez et al., arXiv (2019)

Donnelly et al., Nat. Nano. (2020)
 Witte et al., Nano Lett. (2020)
 Seki et al., Nat. Mat (2021)
 Finizio et al., Nano Lett. (2022)
 Di Pietro et al., PRB (2023)

Spatial Resolution:

< 10 nm

Sample thickness:

< 200 nm

Spatial Resolution:

~10-100 μm

Sample thickness:

up to mms

Spatial Resolution:

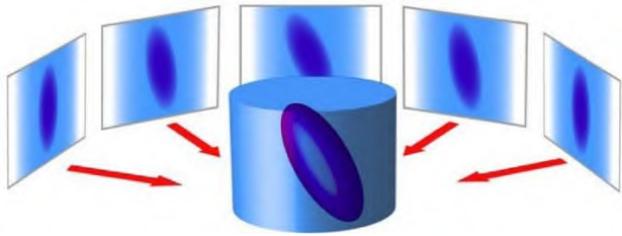
~10 -100 of nm

Sample thickness:

Up to 10s of μm

DEVELOPING 3D MAGNETIC IMAGING

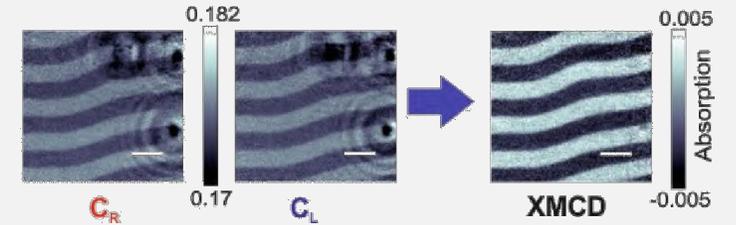
3D imaging: tomography



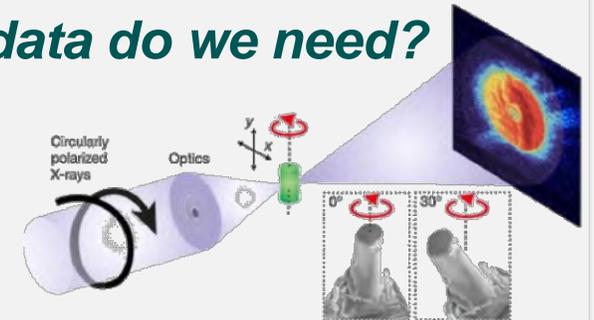
Guizar-Sicairos et al. Optics Express 19, 21345 (2011)

Ingredients of magnetic tomography?

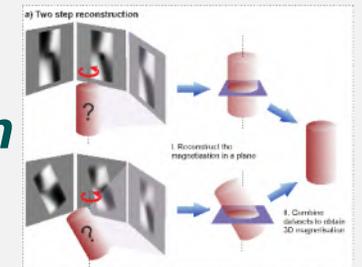
2D X-ray magnetic imaging



What data do we need?

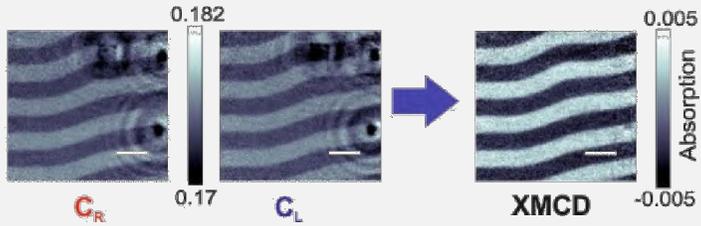


New reconstruction algorithm

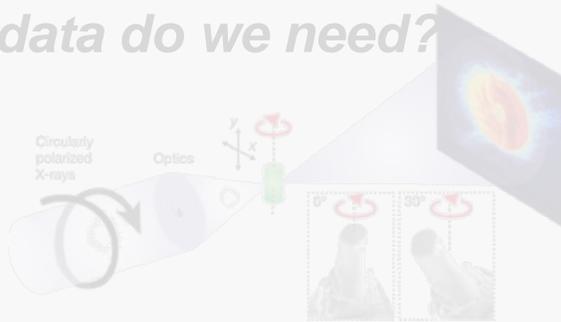


DEVELOPING 3D MAGNETIC IMAGING

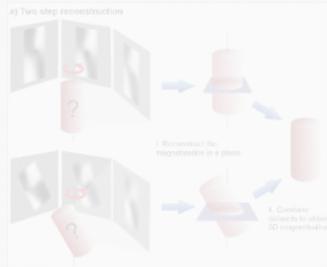
2D X-ray magnetic imaging



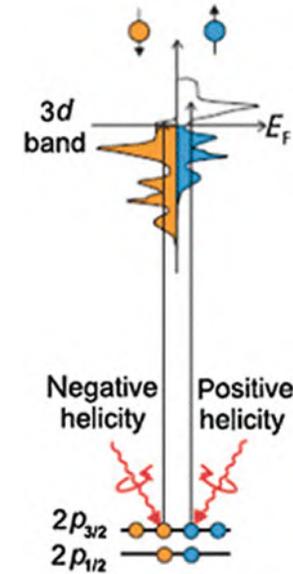
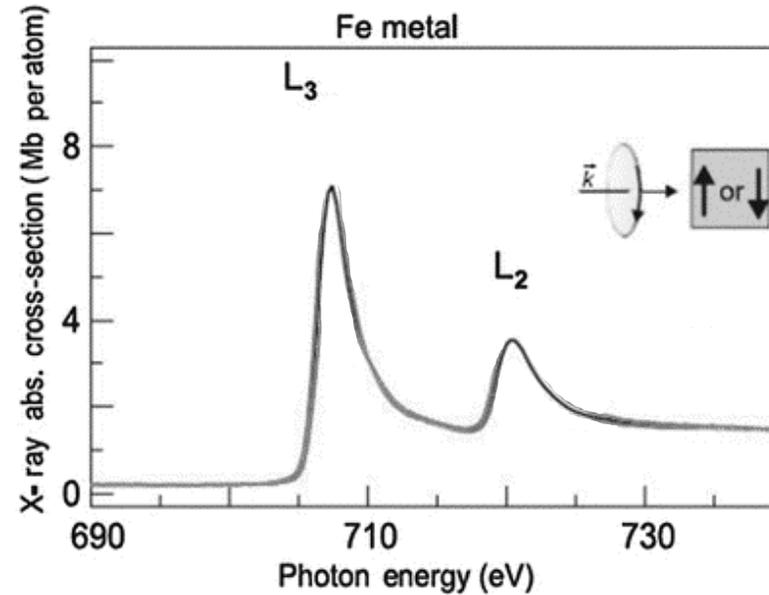
What data do we need?



New reconstruction algorithm



X-ray magnetic circular dichroism:



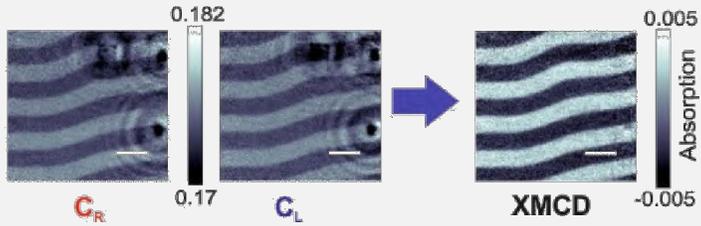
Stohr, Magnetism: from fundamentals to Nanoscale dynamics (2006)

$$f = \underbrace{f_c(\epsilon_f^* \cdot \epsilon_i)}_{\text{“Electronic”}} - \underbrace{if_m^{(1)}(\epsilon_f^* \times \epsilon_i) \cdot \mathbf{m}(\mathbf{r}) + f_m^{(2)}(\epsilon_f^* \cdot \mathbf{m}(\mathbf{r}))(\epsilon_i \cdot \mathbf{m}(\mathbf{r}))}_{\text{“Magnetic”}}$$

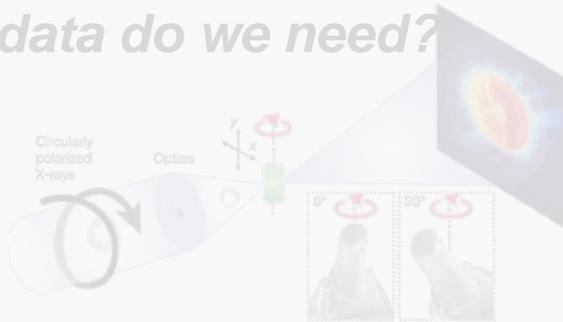
On resonance:
scattering factor dependent
on polarisation and \hat{m} !

DEVELOPING 3D MAGNETIC IMAGING

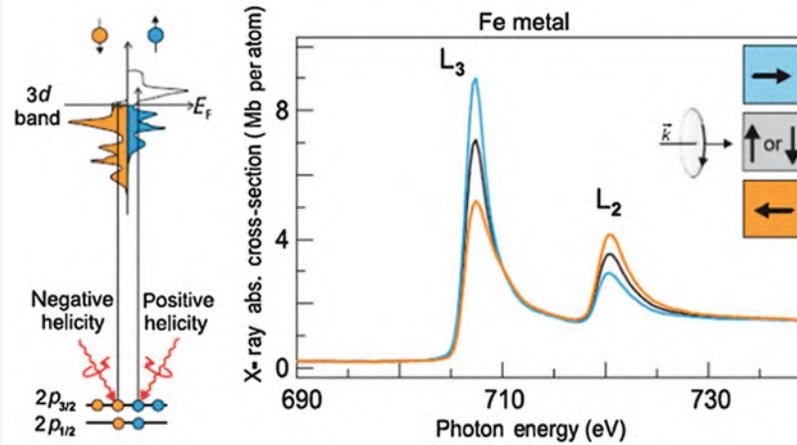
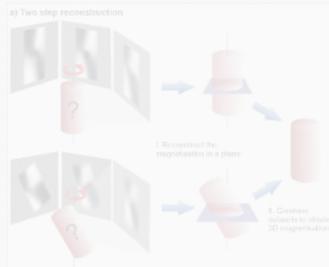
2D X-ray magnetic imaging



What data do we need?



New reconstruction algorithm

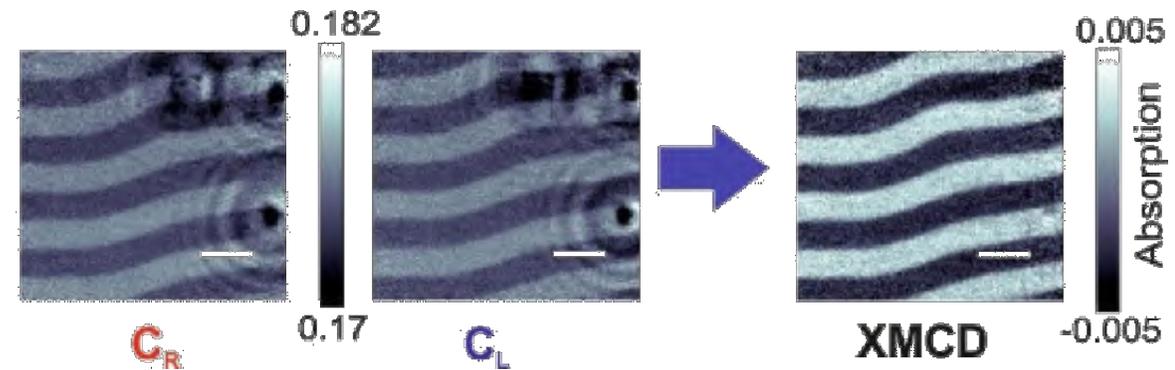


Stohr, Magnetism: from fundamentals to Nanoscale dynamics (2006)

Soft X-rays: directly probe valence band
→ *strong signal*

Hard X-rays: indirectly probe valence band
→ *weak signal*

Combine with microscopy:

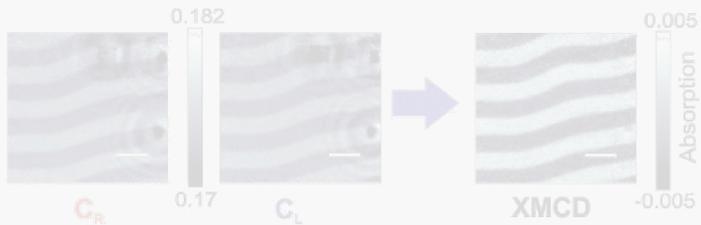


Donnelly et al., Phys. Rev. B **94**, 064421 (2016)

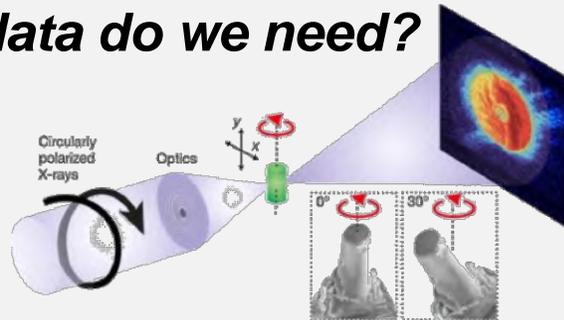


DEVELOPING 3D MAGNETIC IMAGING

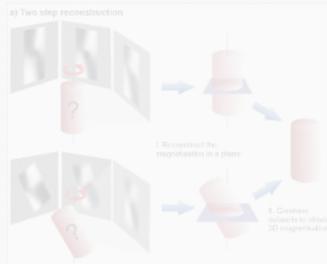
2D X-ray magnetic imaging



What data do we need?



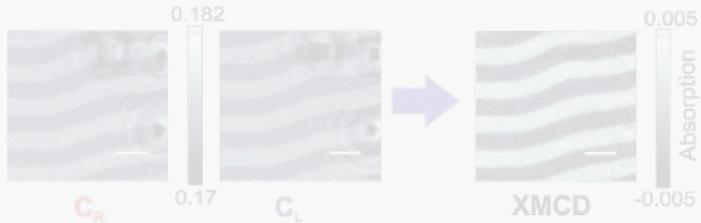
New reconstruction algorithm



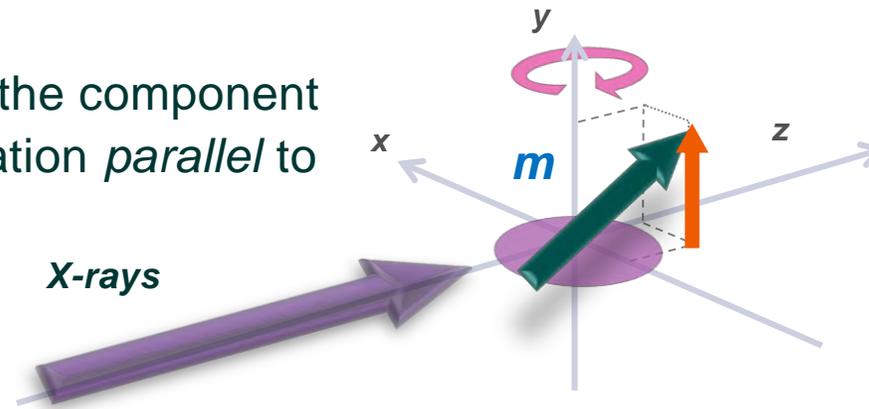
Question!
How many tomograms do we need?

DEVELOPING 3D MAGNETIC IMAGING

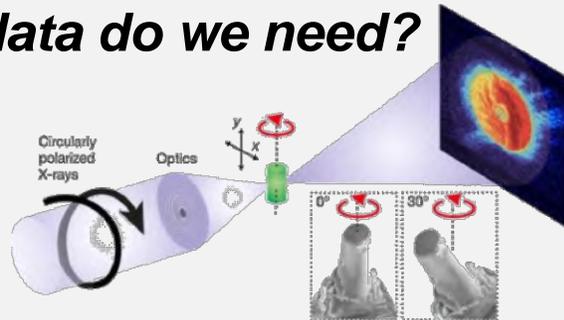
2D X-ray magnetic imaging



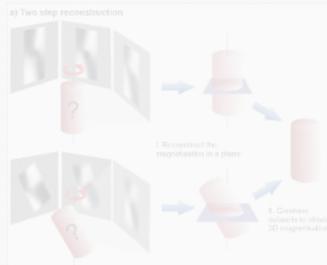
Sensitive to the component of magnetisation *parallel* to the X-rays



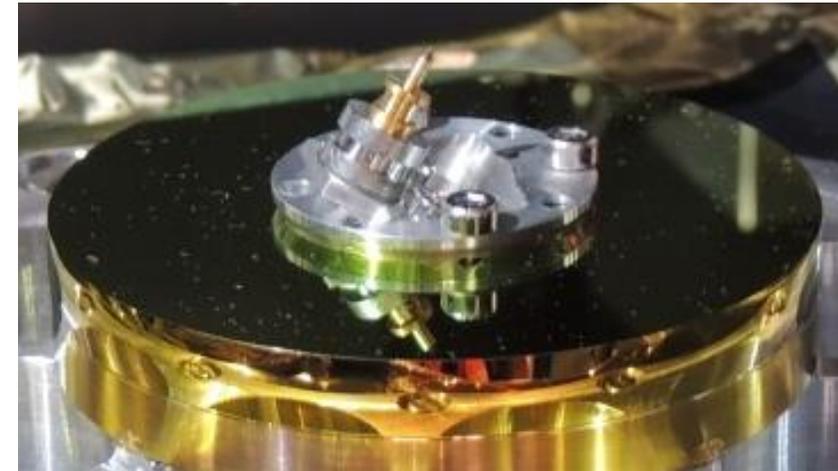
What data do we need?



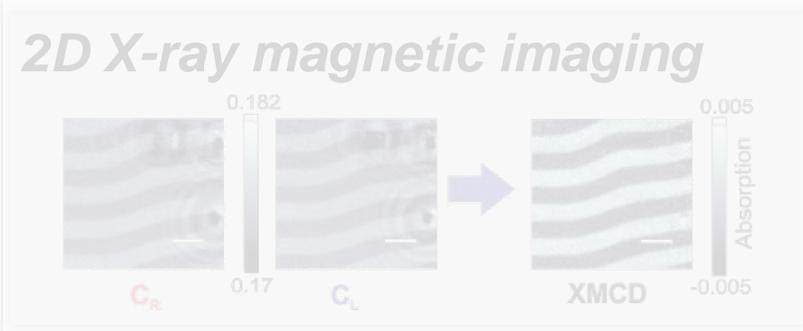
New reconstruction algorithm



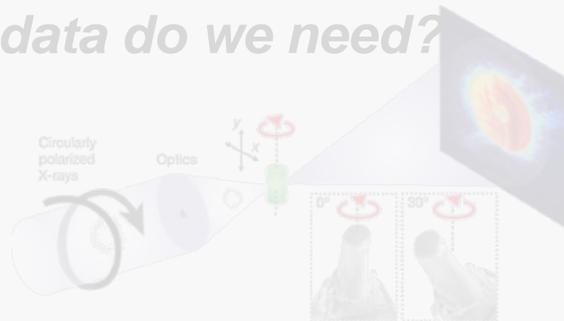
To be sensitive to the *y* component, we tilt the sample and remeasure:



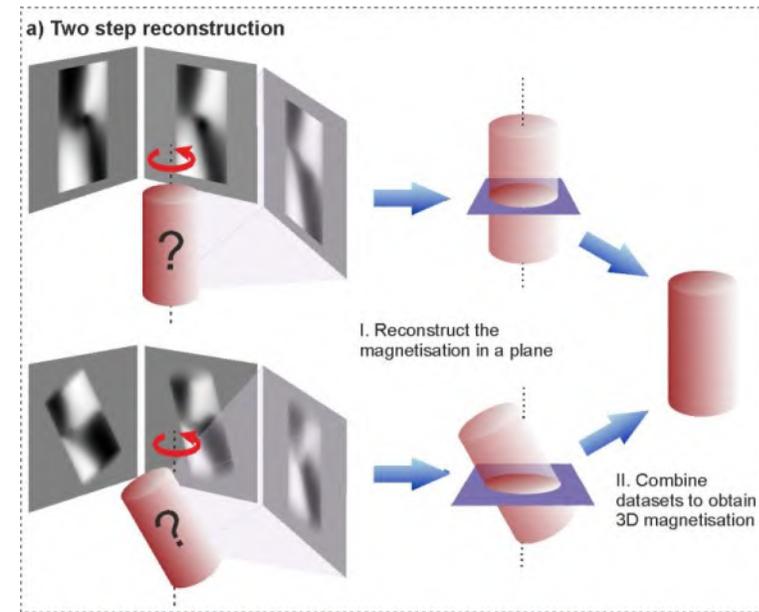
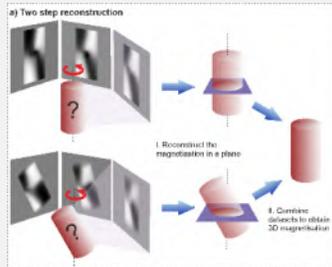
DEVELOPING 3D MAGNETIC IMAGING



What data do we need?



New reconstruction algorithm

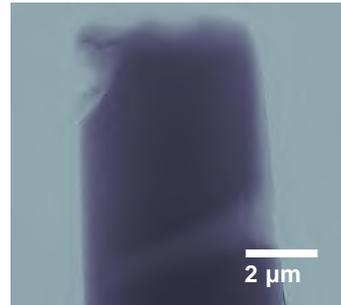


Gradient-based iterative reconstruction algorithm

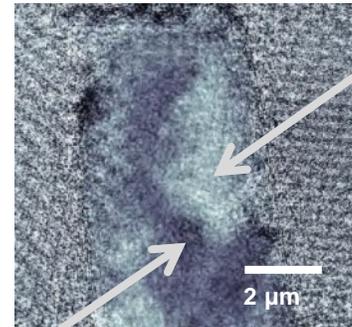


X-RAY MAGNETIC TOMOGRAPHY

Absorption image (C_L)



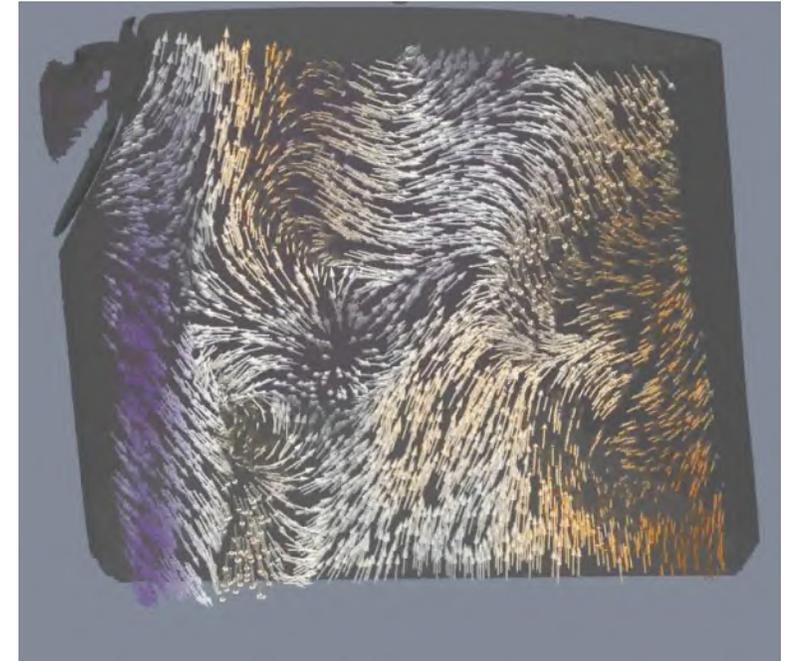
XMCD image ($C_L - C_R$)



Pointing towards us

Pointing away from us

Reconstruct with 100 nm spatial resolution

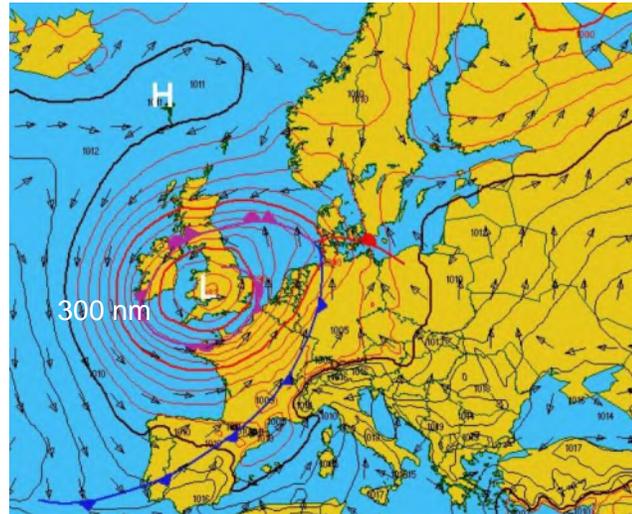
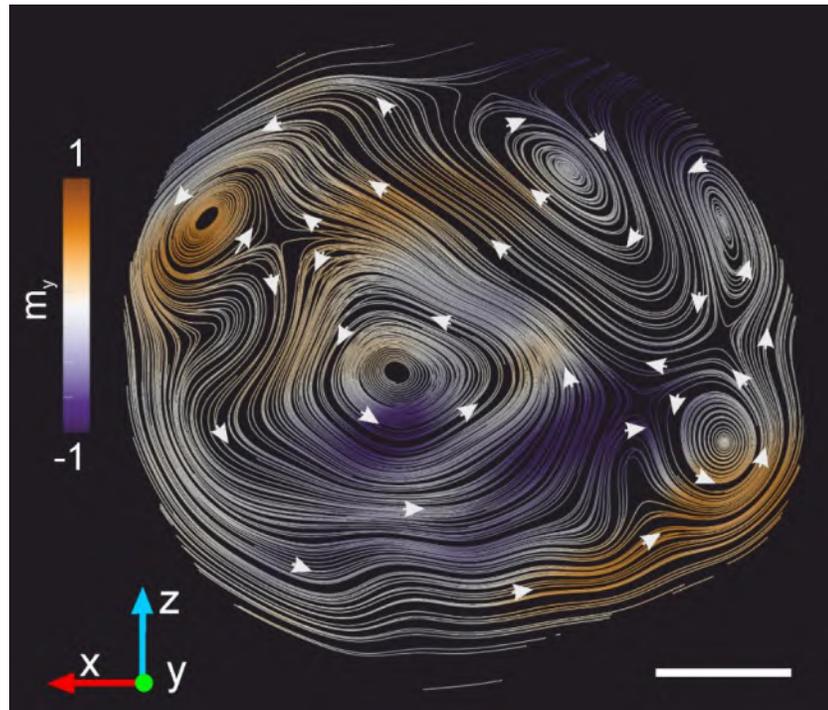


5 μm

Donnelly et al., Nature **547**, 328 (2017)



X-RAY MAGNETIC TOMOGRAPHY



Topology?

Donnelly et al., Nature **547**, 328 (2017)



UNDERSTANDING: TOPOLOGY

Key: *Smoothly deform!*



Meaning:



~



≠

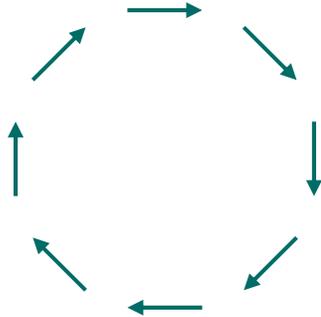
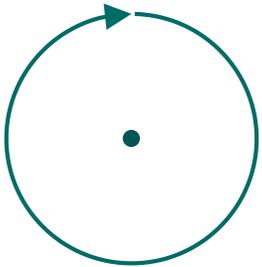


For magnetism?

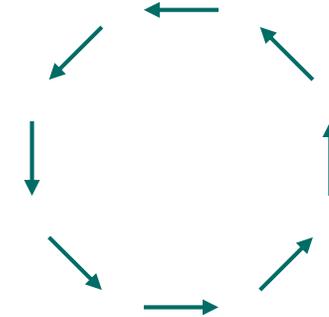
→ *assume continuous vector field*

Let's start with 2D!

Winding number:



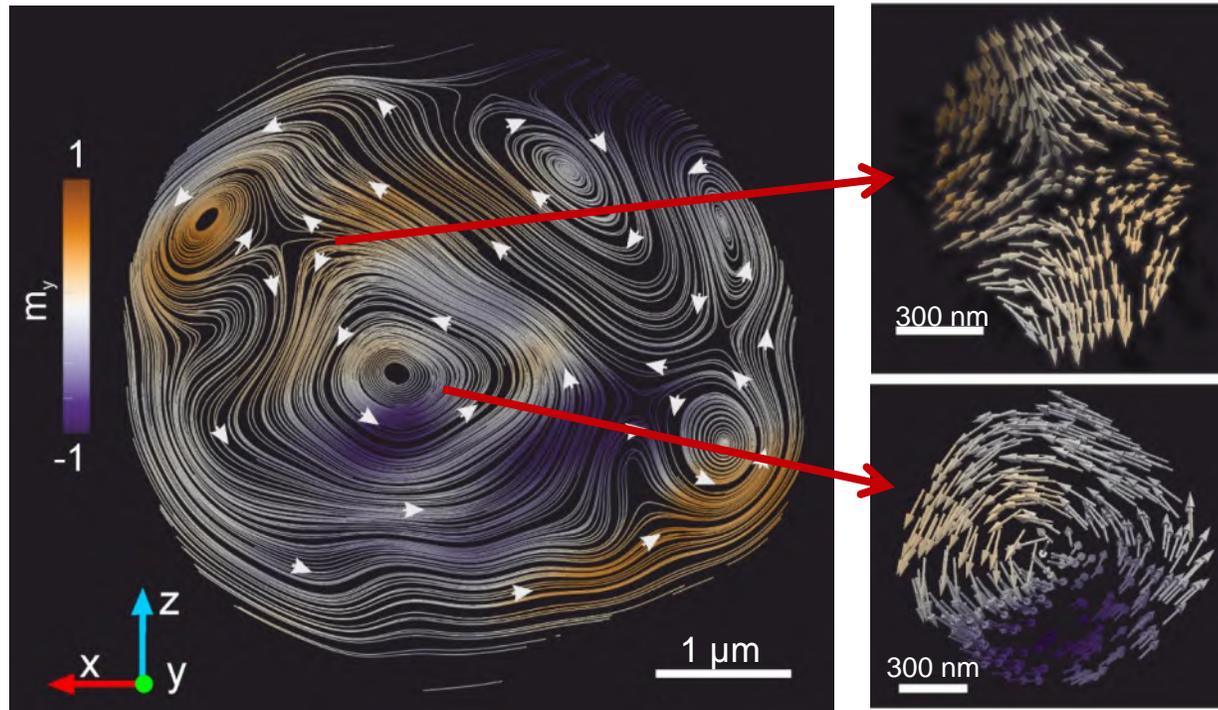
Wind clockwise
→ $W=+1$



Wind clockwise
→ $W=+1$

Are these topologically equivalent?

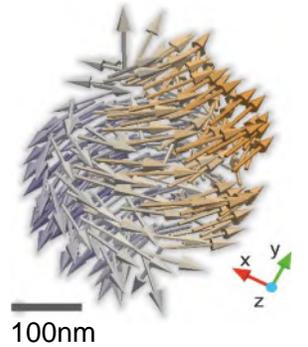
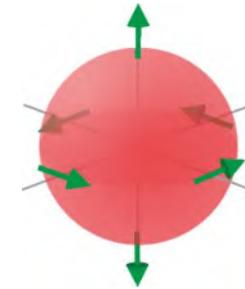
X-RAY MAGNETIC TOMOGRAPHY



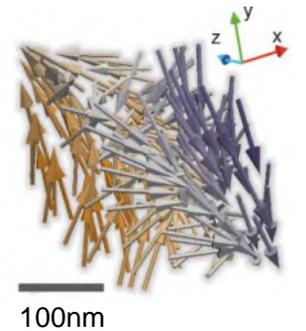
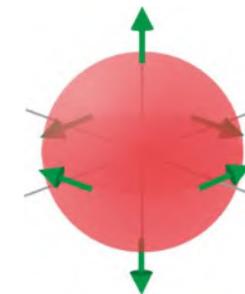
Anti-vortex



Vortex



**Magnetisation
Singularities
*Bloch points***

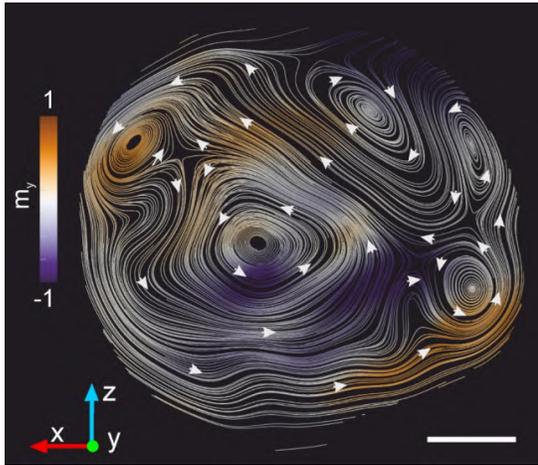


Donnelly et al., Nature **547**, 328 (2017)

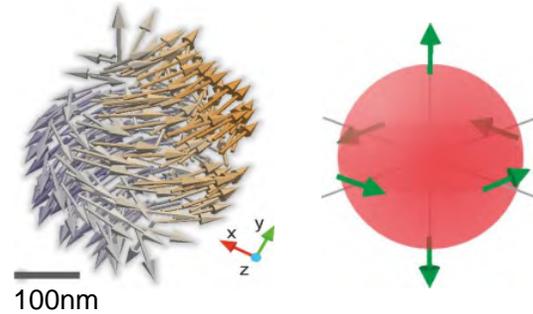


3D MAGNETIC IMAGING

3D vortices & antivortices

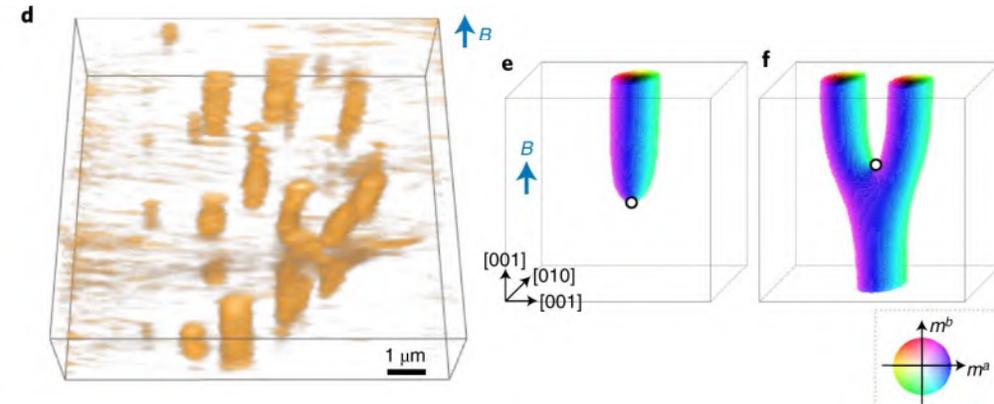


Bloch point singularities



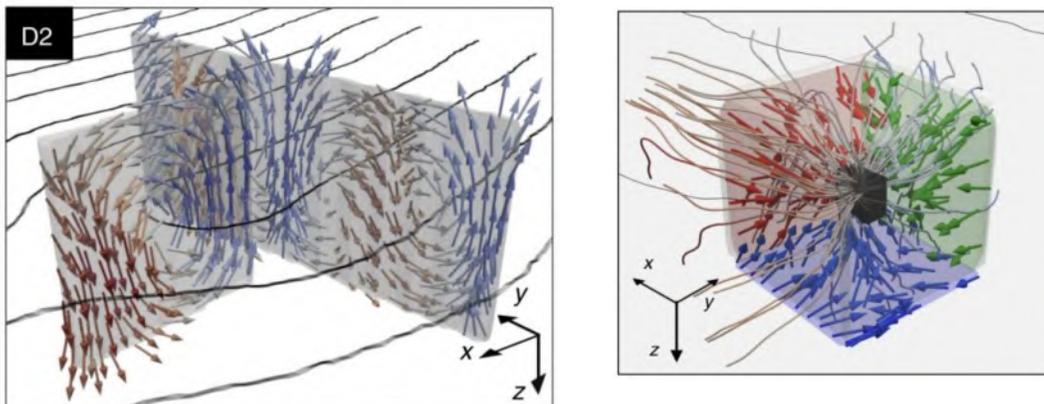
Donnelly et al., Nature 547 328 (2017)

Skyrmions & Bobbers



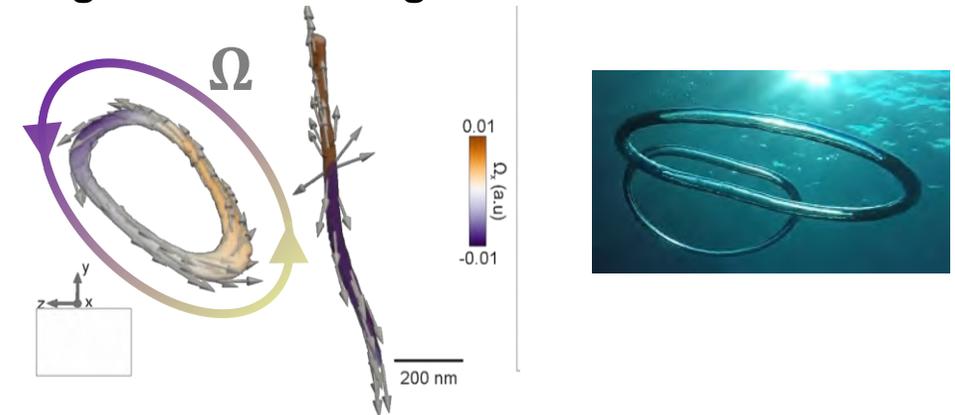
Seki et al., Nature Materials 21, 181 (2022)

Closure domains & topological charge of Bloch points



Hierro Rodriguez et al., Nat. Comm. 11 6382 (2020)

Magnetic vortex rings



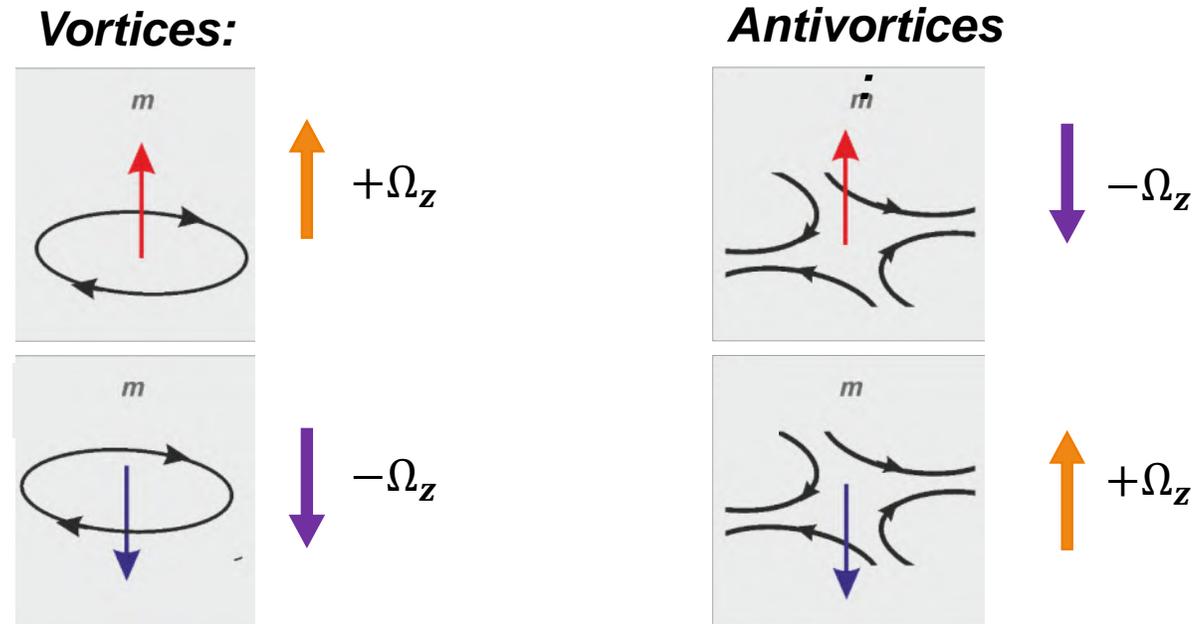
Donnelly et al., Nature Physics 17, 316 (2021)



IMPORTANCE OF DATA ANALYSIS! FOR EXAMPLE: → INSIGHT INTO TOPOLOGICAL STRUCTURES

Through calculations of the magnetic vorticity Ω :

N. Papanicolaou, NATO ASI Series C404, 151-158 (1993).
Cooper, PRL 82 1554 (1999)



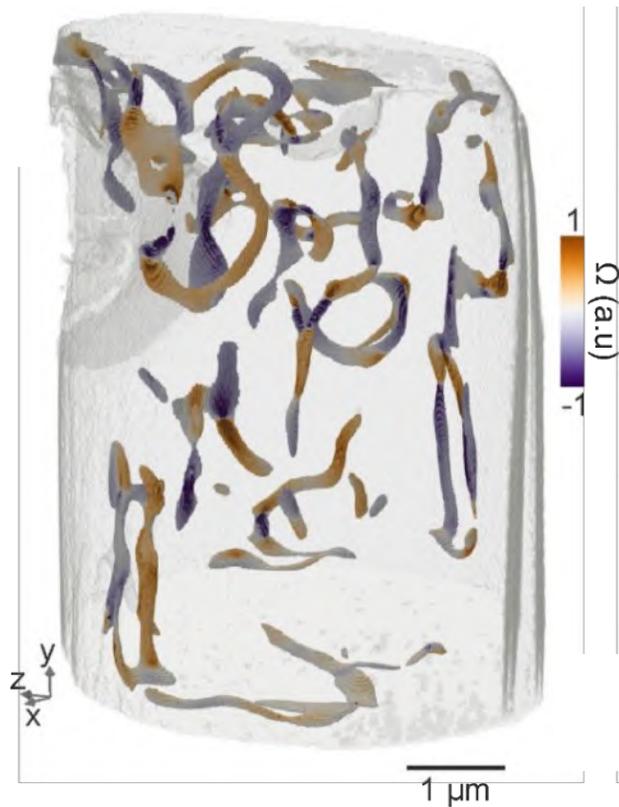
Magnetic vorticity Ω = flux of Skyrmion number density

Donnelly et al., Nature 547, 328 (2017)

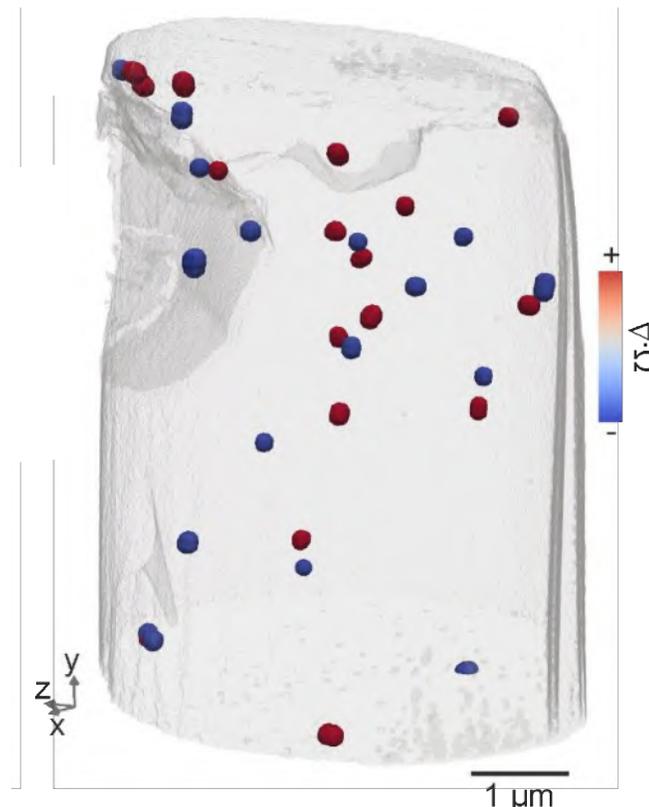
Donnelly et al., Nature Physics 17, 316 (2021)

IMPORTANCE OF DATA ANALYSIS! FOR EXAMPLE: → INSIGHT INTO TOPOLOGICAL STRUCTURES

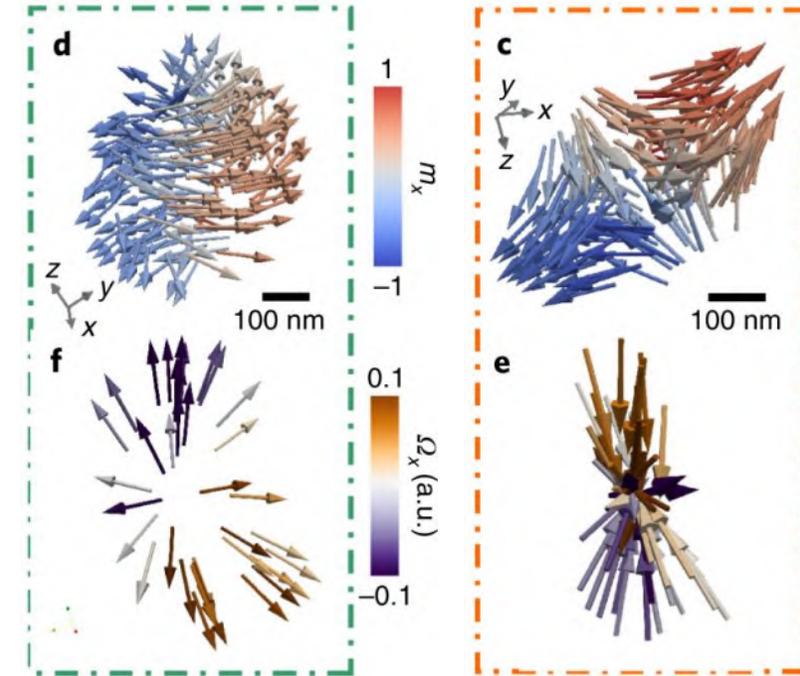
Through calculations of the magnetic vorticity Ω :



**Identify & understand
3D structures**



**Locate singularities
within the bulk**



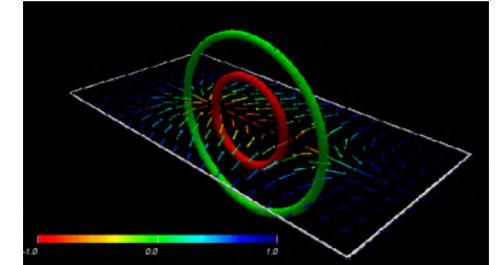
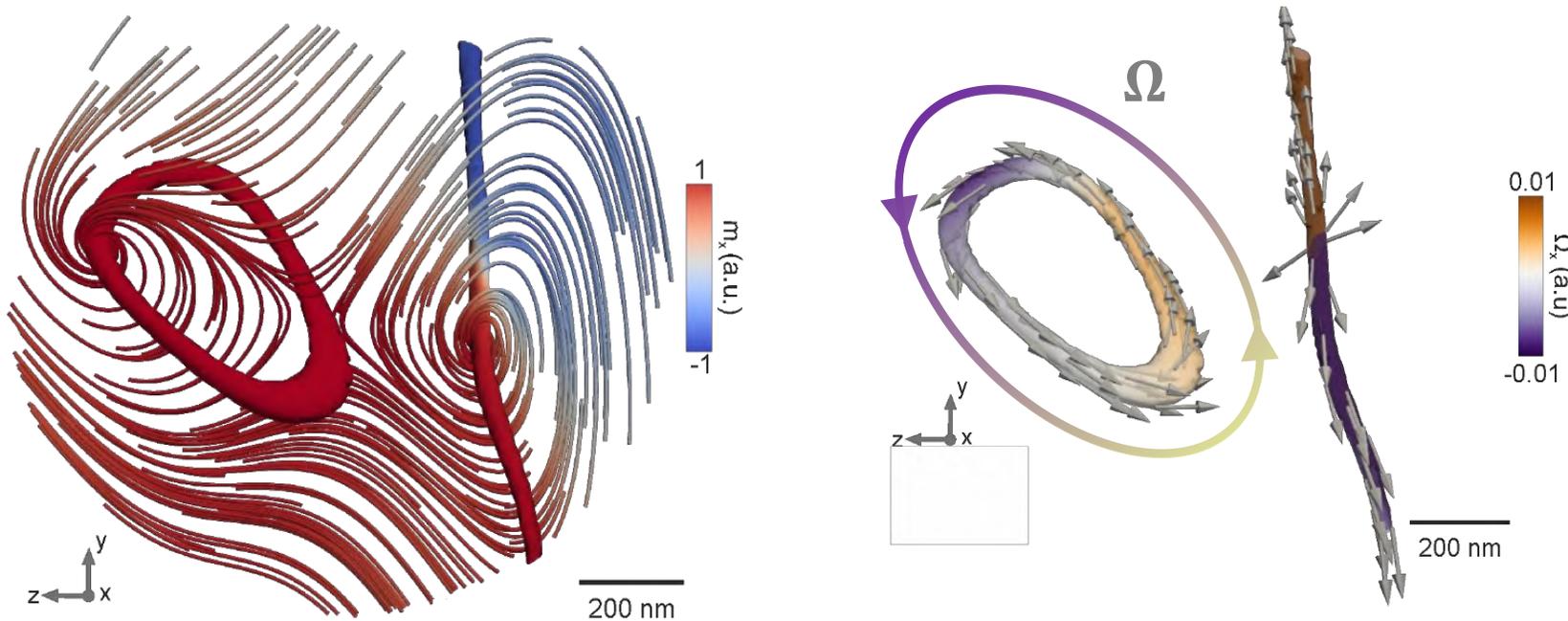
Similar analysis:
Hierro-Rodriguez et al.,
Nature Communications 11 6382 (2020)

Donnelly et al., Nature 547, 328 (2017)
Donnelly et al., Nature Physics 17, 316 (2021)



IMPORTANCE OF DATA ANALYSIS! FOR EXAMPLE: → INSIGHT INTO TOPOLOGICAL STRUCTURES

Unexpectedly stable loops:



Cooper, PRL 82 1554 (1999)

Magnetic vortex rings!



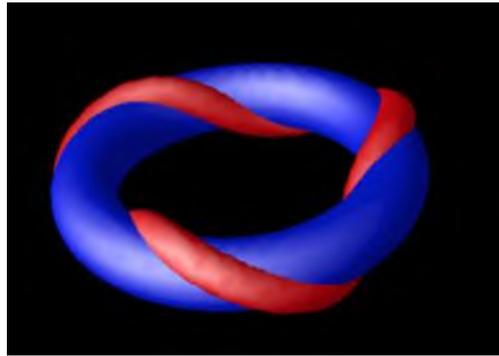
Donnelly et al., Nature Physics 17, 316 (2021)



WE CAN USE THIS TO UNDERSTAND NEW 3D TEXTURES:

Could it be the topology of the vortex ring?

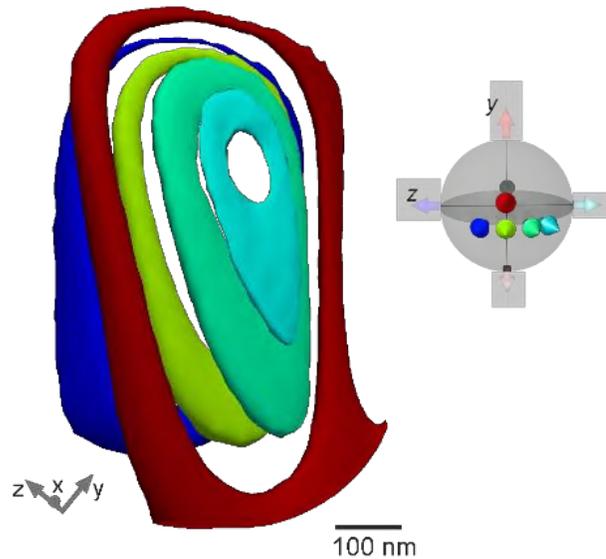
By plotting “pre-images” in 3D:



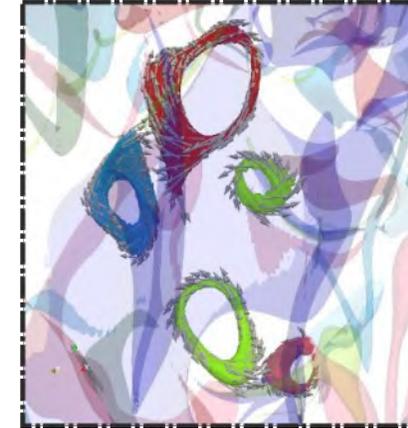
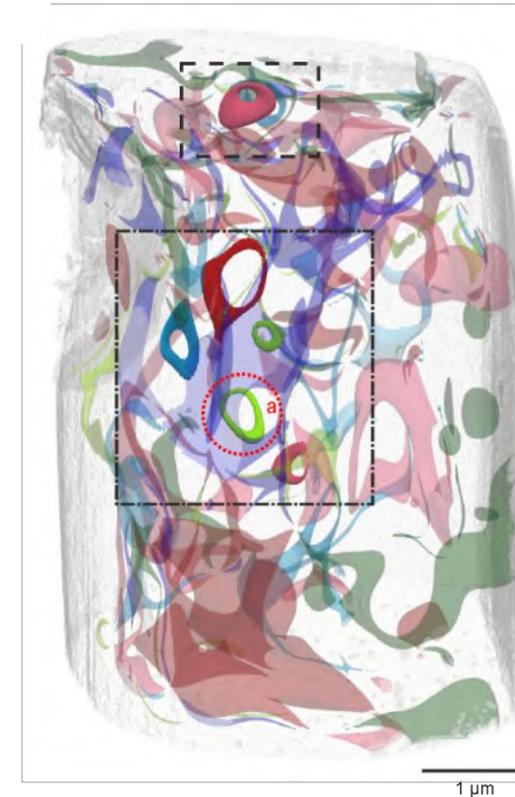
$$-\frac{1}{\sqrt{2}}(\hat{y} + \hat{z})$$

Sutcliffe PRL 118, 247203 (2017)

Pre-images link 3 times
→ **Hopf Index H=3**



Pre-images don't link
→ **Hopf Index H=0**

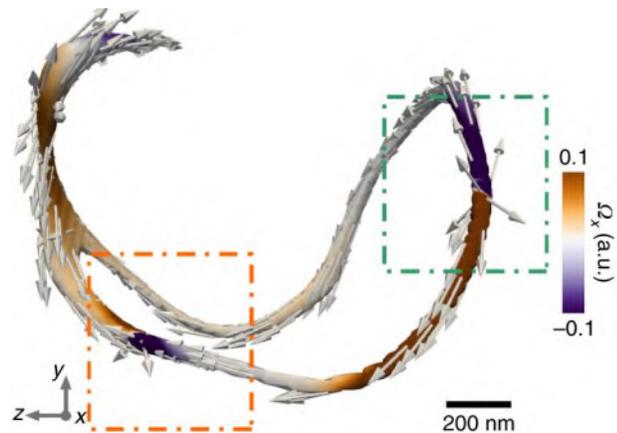
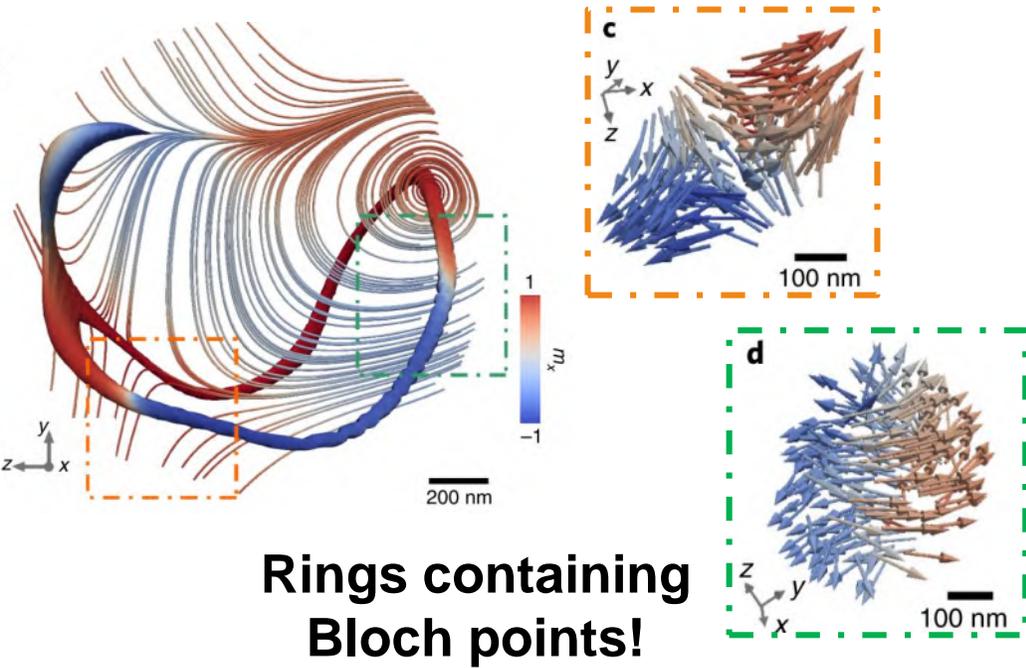


**Multiple vortex rings
→ magnetostatics key!**

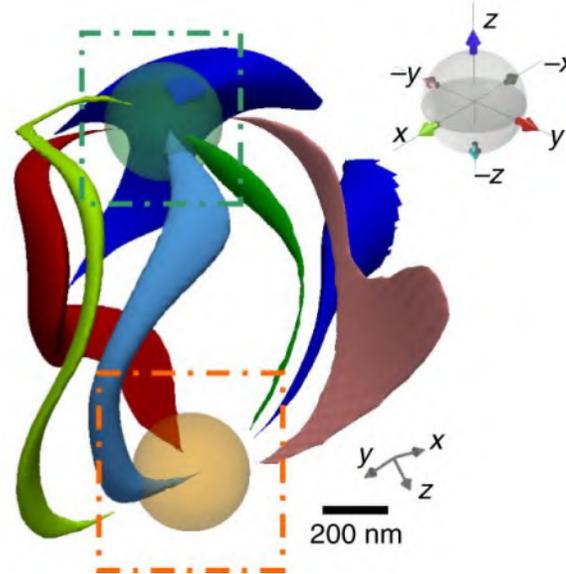
Donnelly et al., Nature Physics 17, 316 (2021)



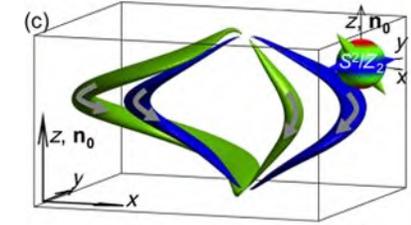
BEYOND SIMPLE VORTEX RINGS:



Plotting pre-images:



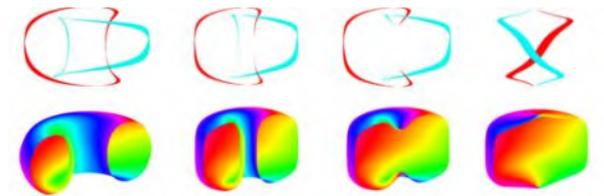
Observed in liquid crystals alongside hopfions



Ackerman et al., PRX 7 011006 (2017)

**Onion shape
→ toron!**

Predicted to be stable alongside hopfions in ferromagnets:



Liu et al., PRB 98 174437 (2018)

Donnelly et al., Nature Physics 17, 316 (2021)

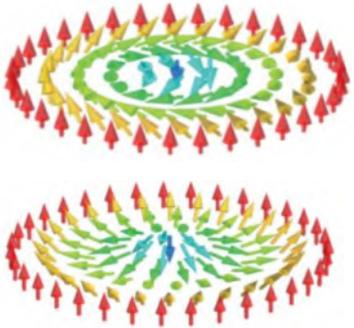


GEOMETRICAL TUNING OF MAGNETIC PROPERTIES

Chirality: DMI

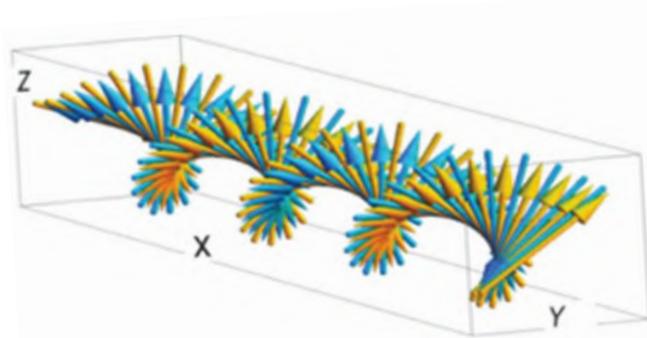
Results in exotic chiral states:

Skyrmions



Fert, et.al. Nat Rev Mat 2017

Spin Spirals

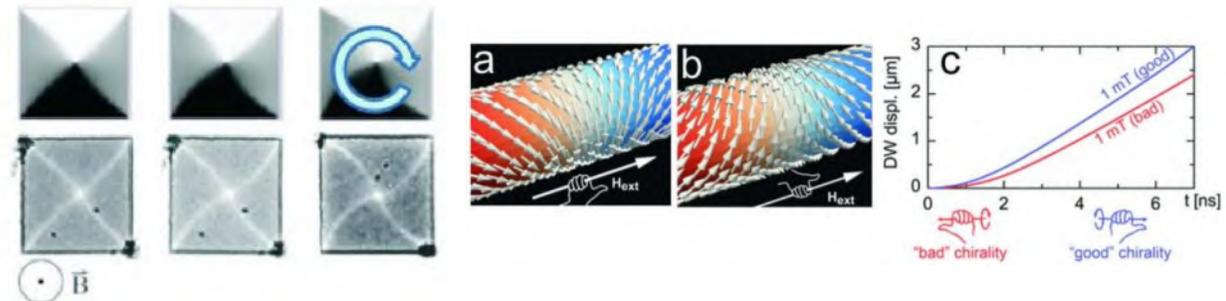


Vedmedenko et al, Phys. Rev. Lett., 2014

Until now, mostly required **specific** materials & interfaces

But...

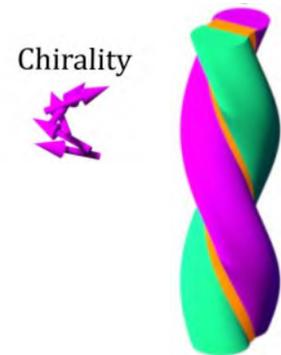
Chirality can also be induced via curvature:



Dietrich et al., PRB 77 174427 (2008)

Hertel, Spin 3 1340009 (2013)

And chiral geometries:

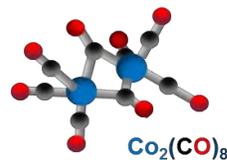
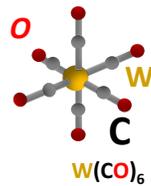
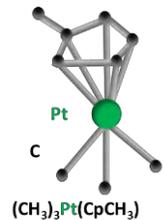
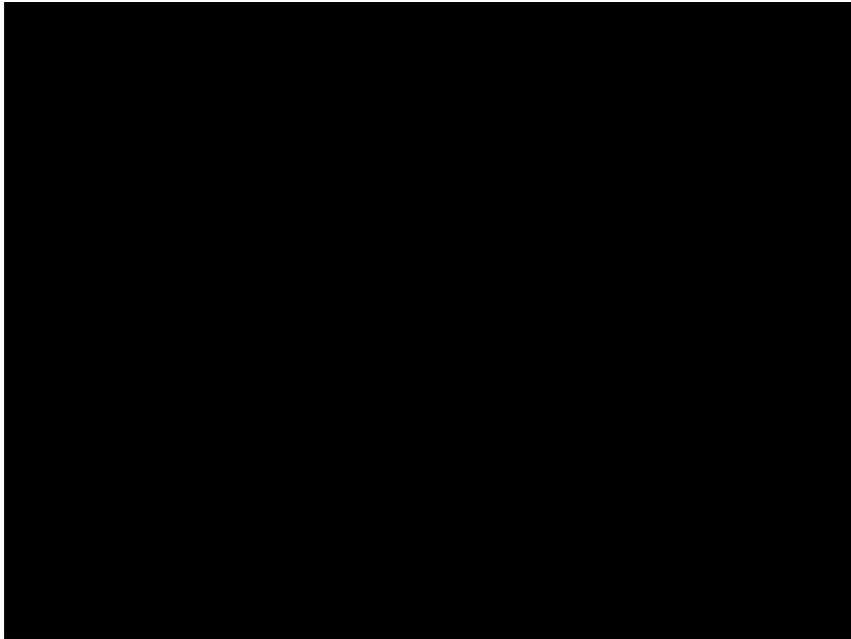




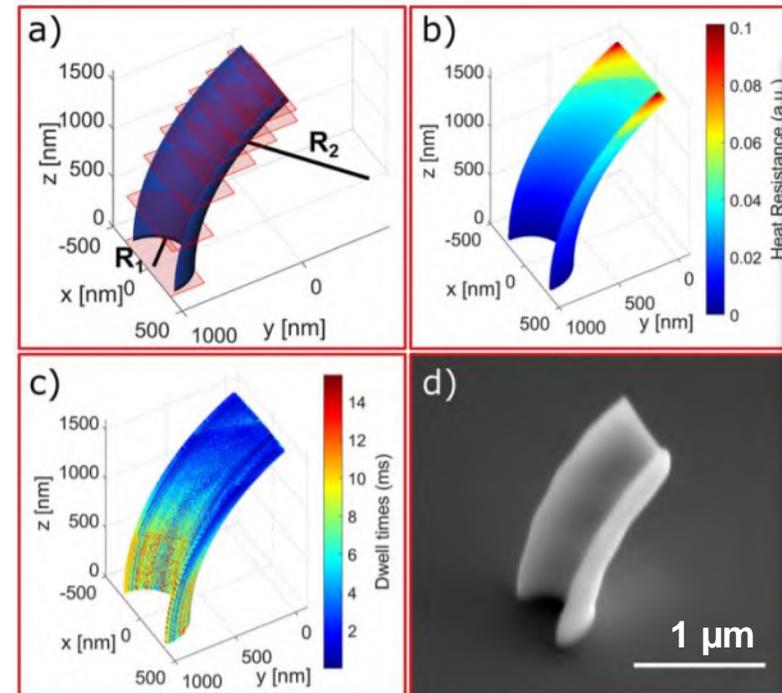
APPLY THIS TO 3D NANOSTRUCTURES?

First: we need to fabricate!

→ Focused electron beam induced deposition



With CAD designs and a growth model:



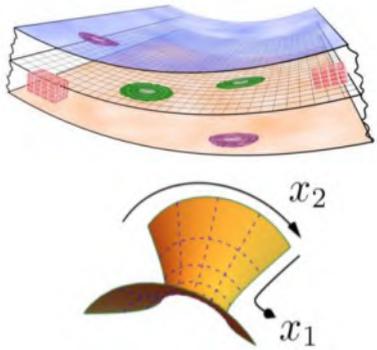
Tuesday
12-sep.
Advanced Fabrication
Denys Makarov
Advanced k-space instrumentation (scatter. & photoemission)
Nicolas Jaouen

Skoric et al., Nano Lett. 20, 184 (2020)

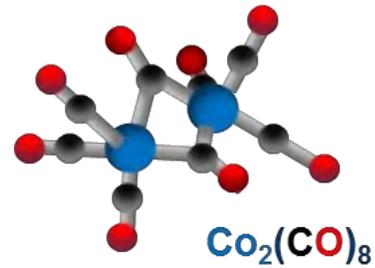


APPLY THIS TO 3D NANOSTRUCTURES?

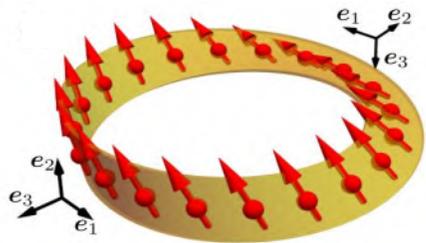
Curved surfaces



Sheka et al., *Comm. Phys.* **3**, 1 (2020).



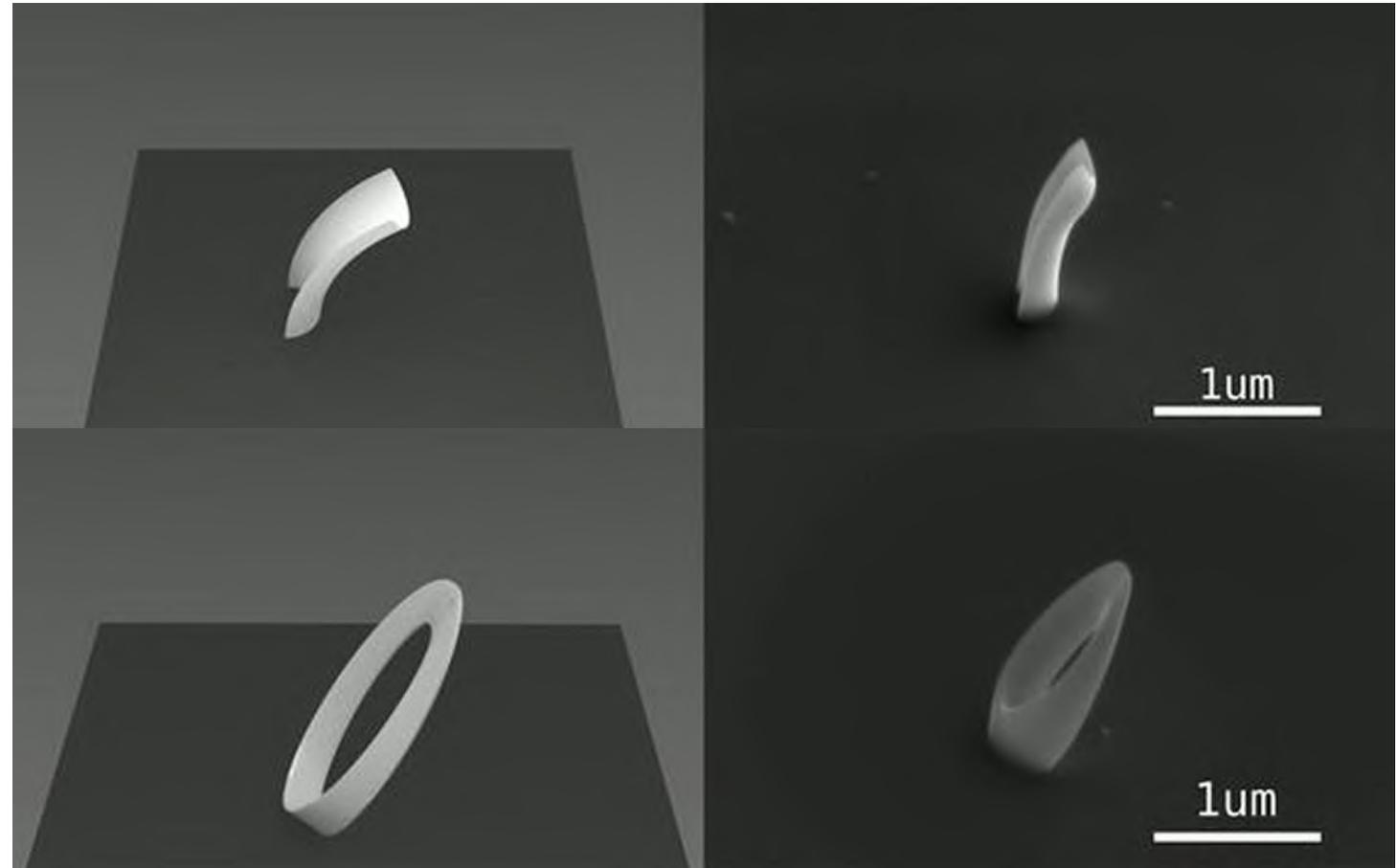
Topological objects



Gaididei et al., *J. Phys. A: Math. Theor.* **50**, 385401 (2017).

3D model

Fabricated structure



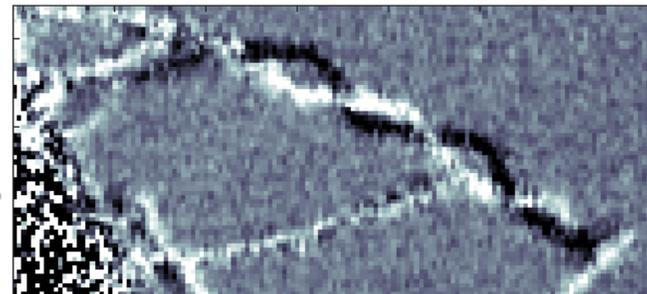
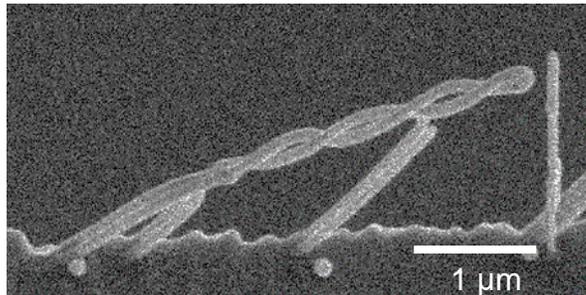
Skoric et al., *Nano Lett.* **20**, 184 (2020)

INFLUENCE OF 3D GEOMETRY ON DOMAIN WALLS

Separated:
Dipolar coupled

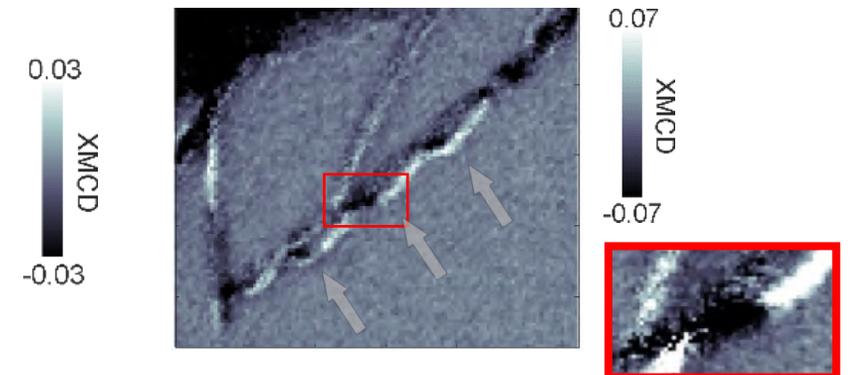


In the as-grown state:



Anti-parallel helices

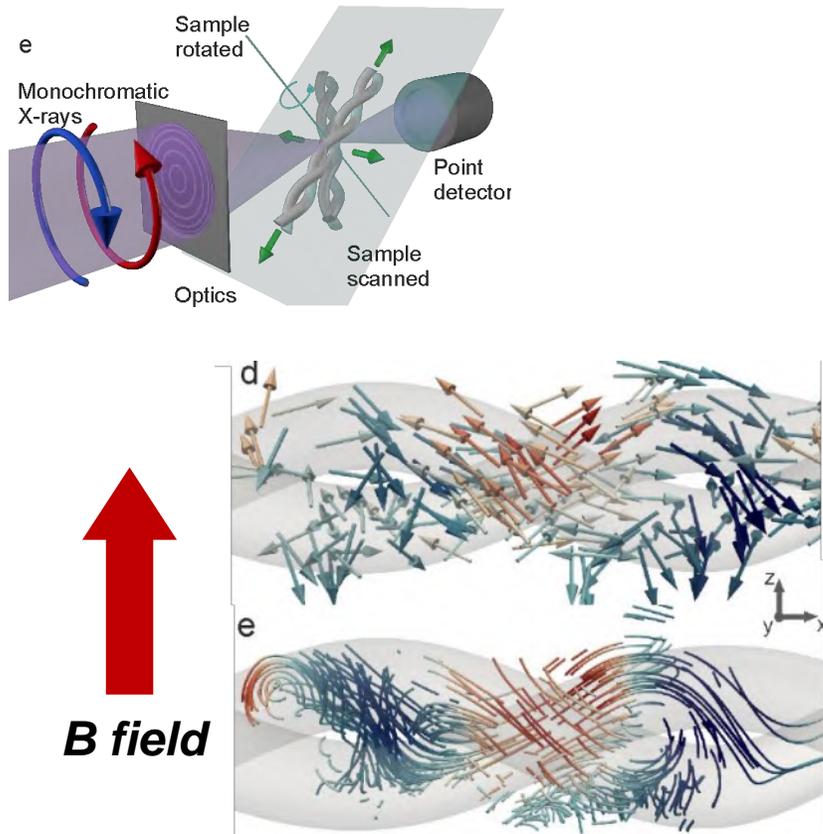
After applying a magnetic field:



Trap domain walls...

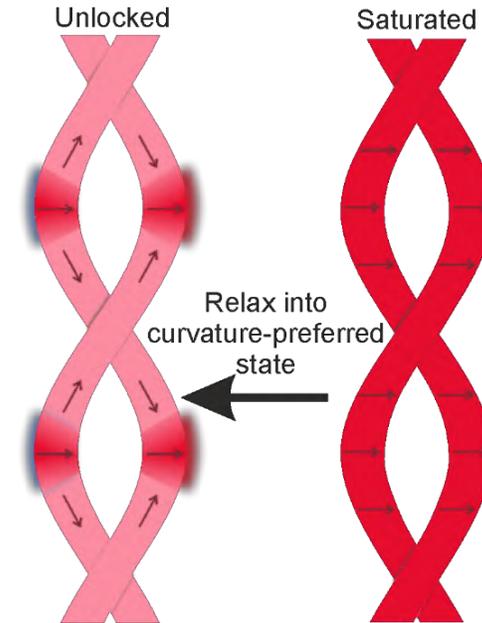
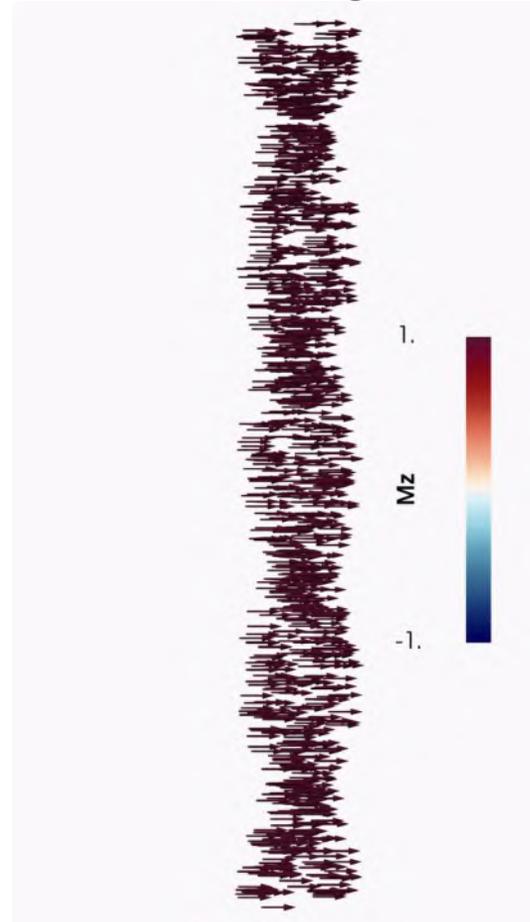
AND IF WE LOOK IN 3D?

Image 3D configuration:



Domain walls appear to have reversed...

Micromagnetic simulations: Magnum.fe



**Interhelix coupling overcomes curvature-induced DMI
→ Locked domain wall state**

Donnelly et al., Nature Nanotechnology 17,136 (2022)



OUR QUESTIONS FOR TODAY:

Higher dimensional investigations?

Vectorial imaging



Magnetisation dynamics

Advanced sample environments

Varying

Temperature,

Pressure,

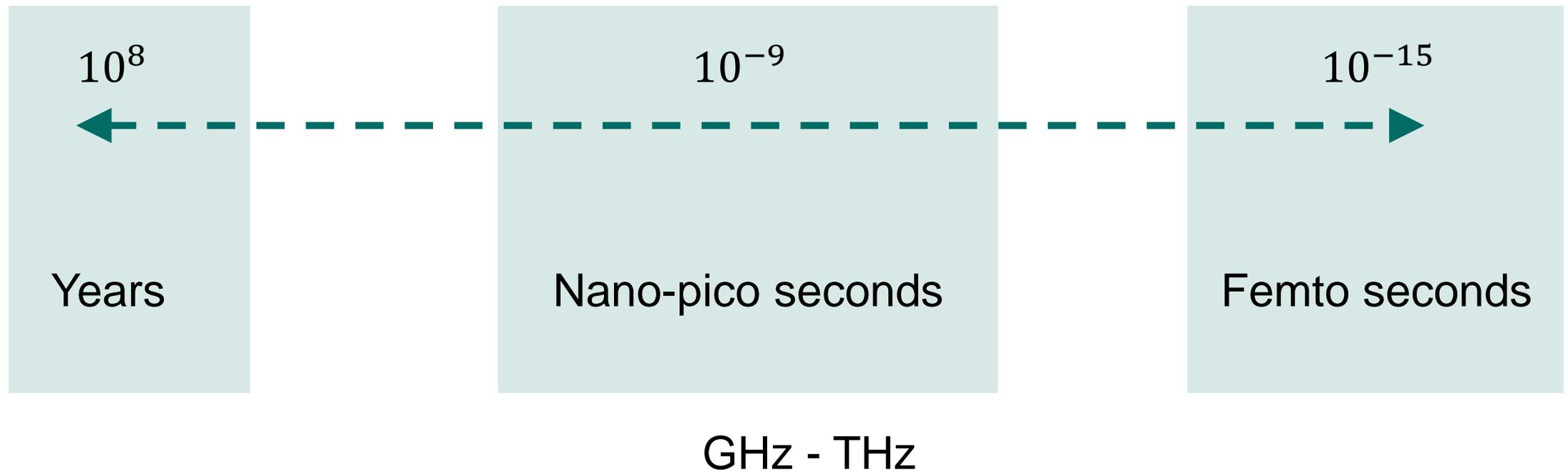
Magnetic field...



DYNAMICS – WHAT TIME SCALES?

Slow!

Ultrafast!





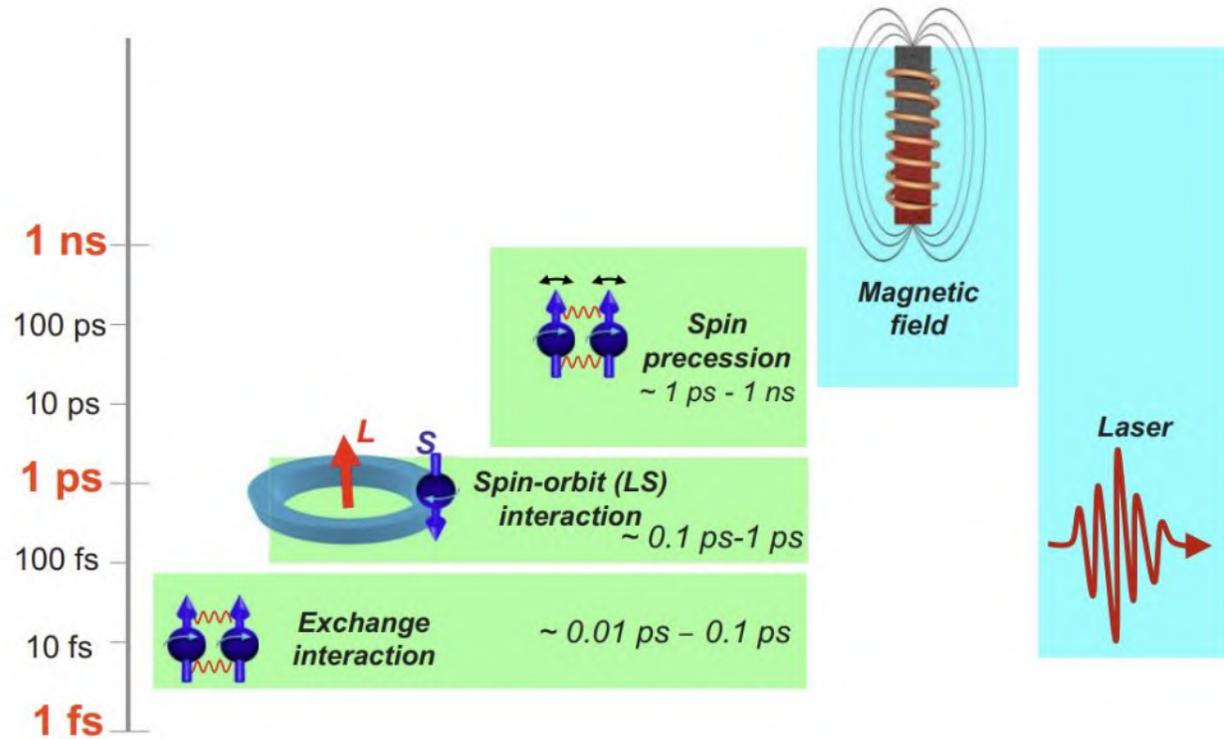
DYNAMICS – WHAT TIME SCALES?

REVIEWS OF MODERN PHYSICS, VOLUME 82, JULY–SEPTEMBER 2010

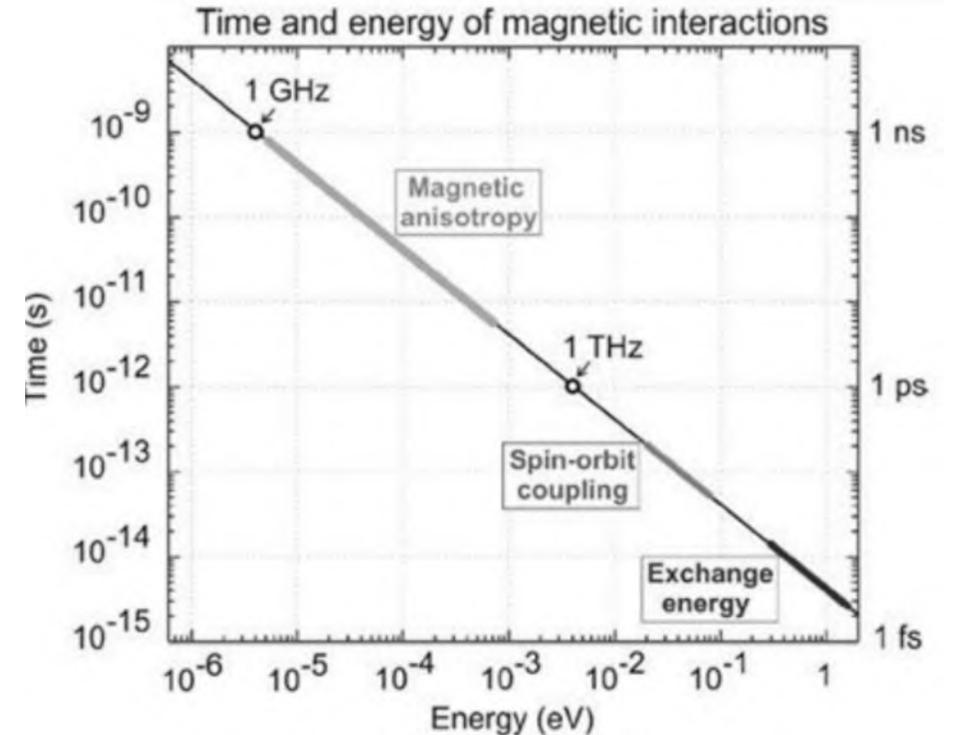
Ultrafast optical manipulation of magnetic order

Andrei Kirilyuk,* Alexey V. Kimel, and Theo Rasing

Radboud University Nijmegen, Institute for Molecules and Materials, Heyendaalseweg
135, 6525 AJ Nijmegen, The Netherlands



There are faster time-scales out there!



Stoehr et al., *Magnetism* Springer-Verlag, (2006)

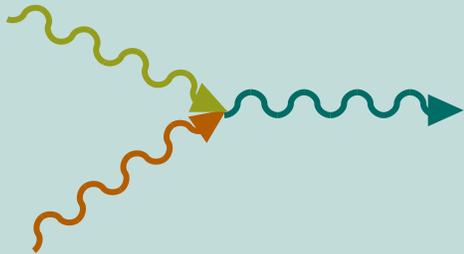


TYPES OF DYNAMIC MEASUREMENTS

Brillouin Light Scattering (BLS)

Spin waves = presence of magnons

Information on spin wave dynamics



Single-shot

Measurement sufficiently short to measure relevant dynamics

Need enough statistics!

Need to measure fast enough

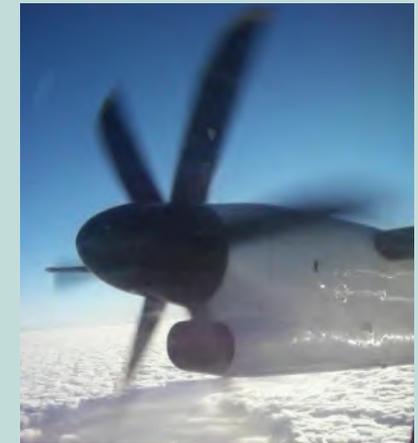


<https://bda668.wordpress.com/>

Pump-probe

Stroboscopic measurements

Look at repetitive processes



https://en.wikipedia.org/wiki/Stroboscopic_effect



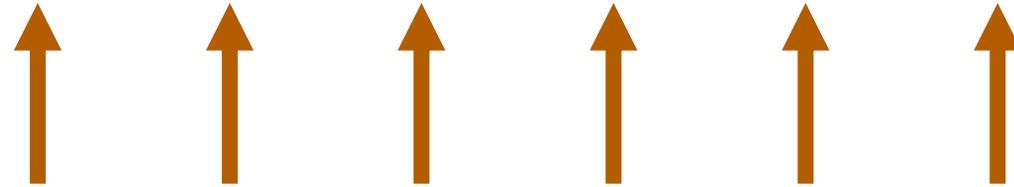
SPIN WAVES: SPIN EXCITATIONS

Brillouin Light Scattering (BLS)

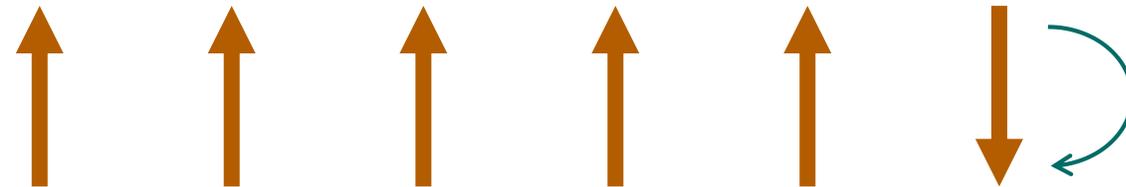
Spin waves = presence of magnons

Information on spin wave dynamics

We have the ground state:

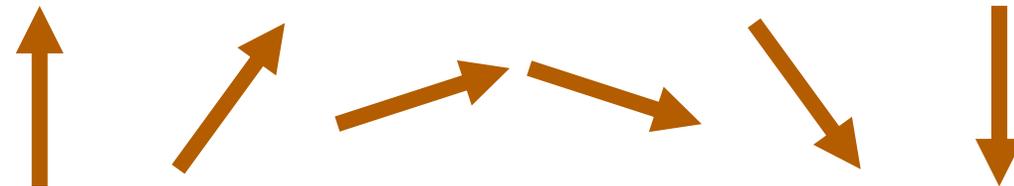


What is the smallest excitation of this ground state?



Flip a single spin

Can distribute over many spins:





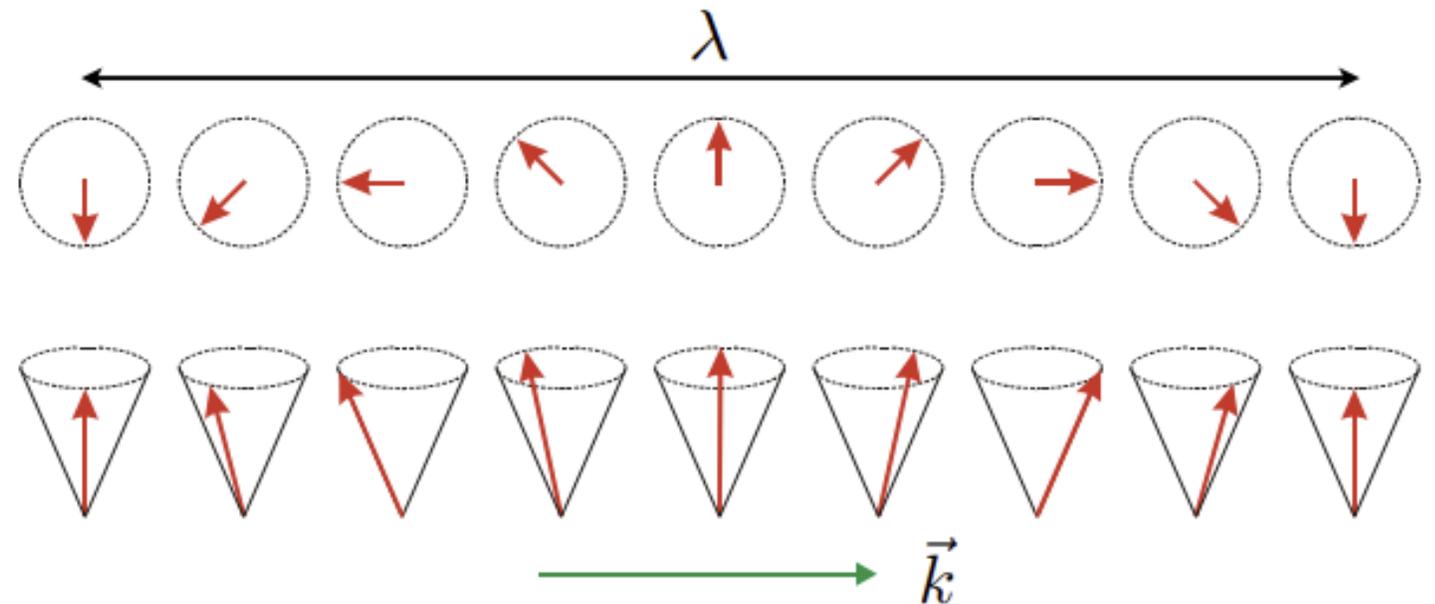
SPIN WAVES

What this leads to:

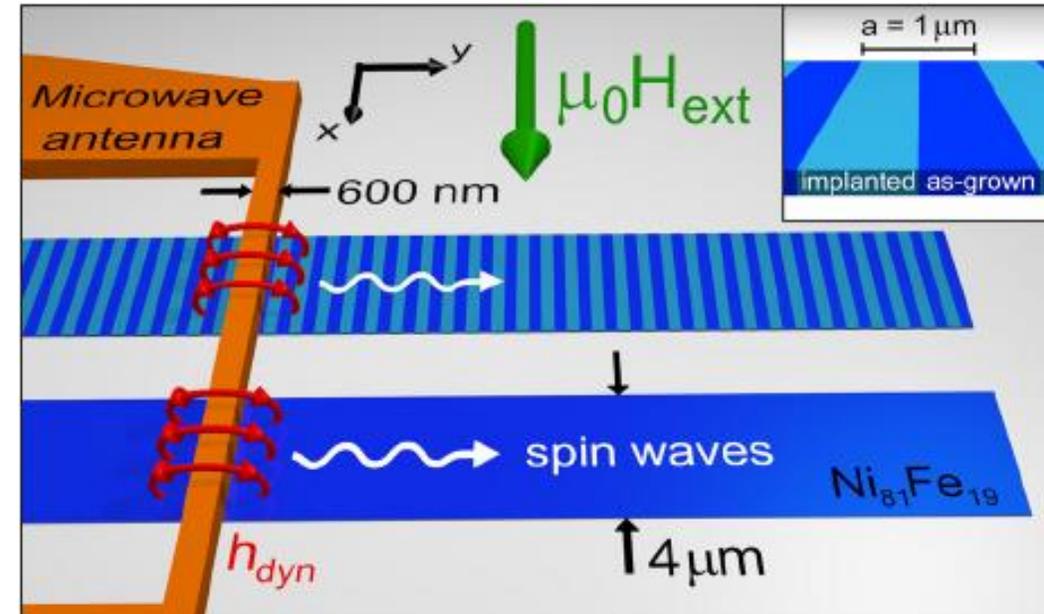
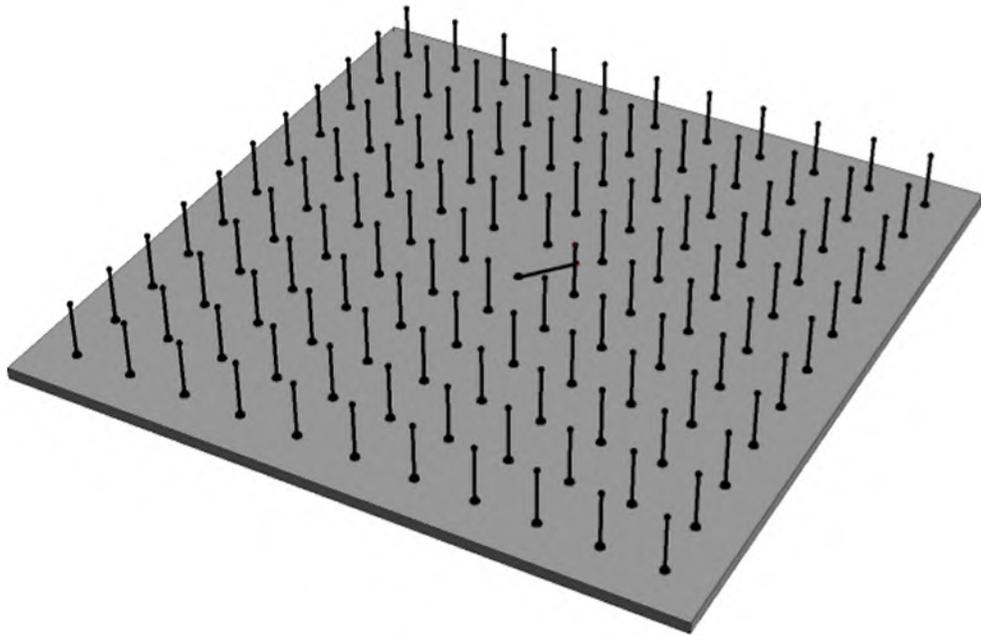
Spins all precessing at the same frequency

But: out of phase with one another:

→ Propagating spin waves



SPIN WAVES: HOW TO EXCITE?



Excite spin waves with an antenna

MEASURING SPIN WAVES

Brillouin Light Scattering (BLS)

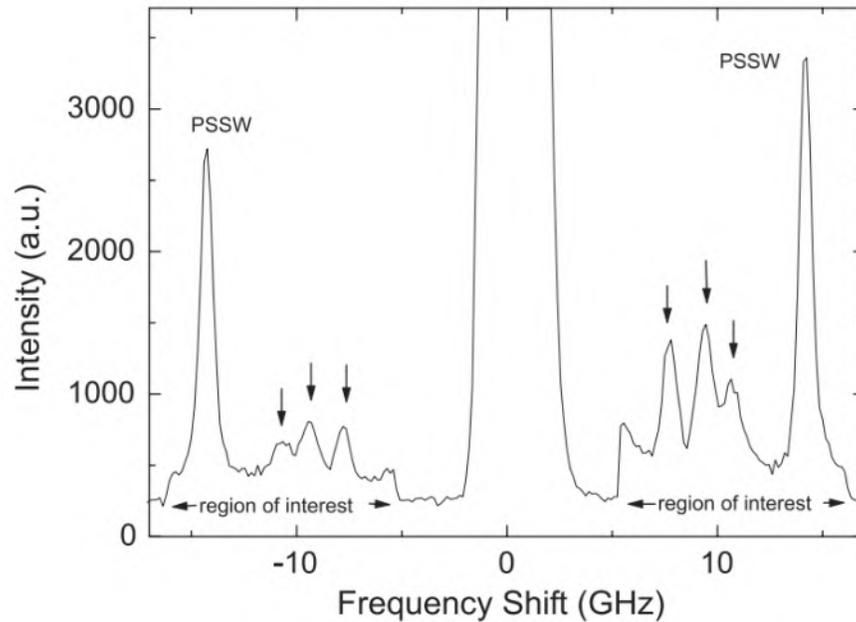
Spin waves = presence of magnons

Information on spin wave dynamics

Brillouin Light Scattering

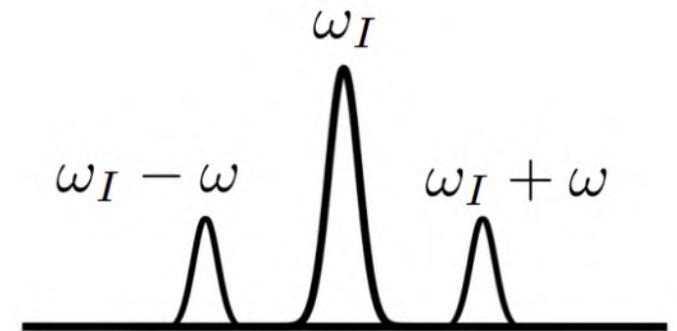
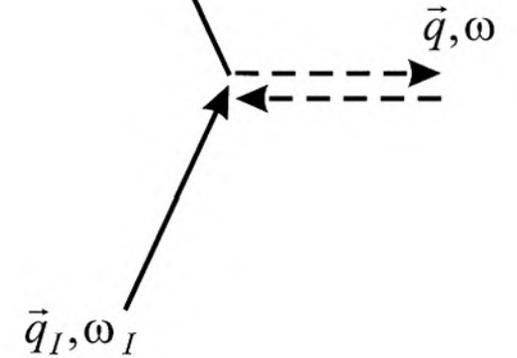
Scatter photons from surface spin waves of films

Reflected photons give information on spin waves that are created (Stokes) or annihilated (anti-Stokes)



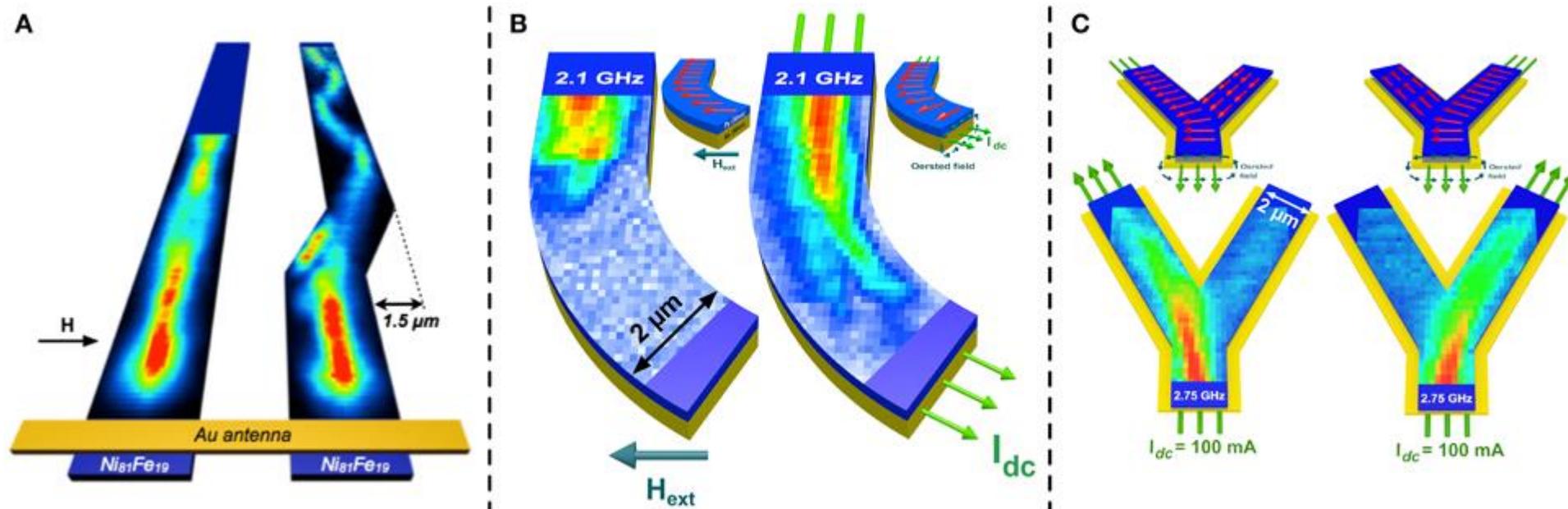
$$\vec{q}_S = \vec{q}_I \pm \vec{q}$$

$$\omega_S = \omega_I \pm \omega$$



MEASURING SPIN WAVES

Brillouin Scattering – microfocused!



Not limited to coherent spin waves!

Sebastian et al., <https://doi.org/10.3389/fphy.2015.00035>

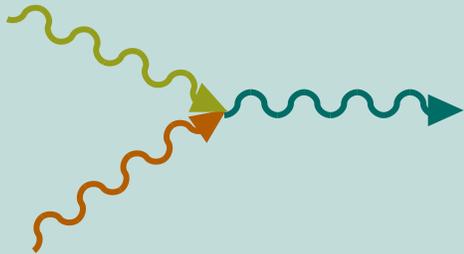


TYPES OF DYNAMIC MEASUREMENTS

Brillouin Light Scattering (BLS)

Spin waves = presence of magnons

Information on spin wave dynamics



Single-shot

Measurement sufficiently short to measure relevant dynamics

Need enough statistics!

Need to measure fast enough

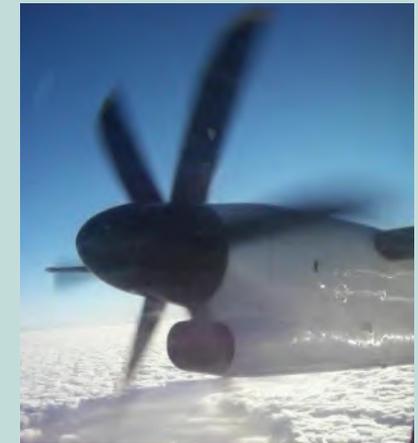


<https://bda668.wordpress.com/>

Pump-probe

Stroboscopic measurements

Look at repetitive processes



https://en.wikipedia.org/wiki/Stroboscopic_effect



IN SITU MEASUREMENTS: CHANGING THE EXCITATION

Single-shot

Measurement sufficiently short to measure relevant dynamics

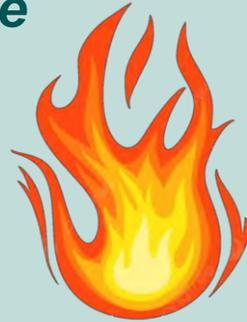
Need enough statistics!

Need to measure fast enough

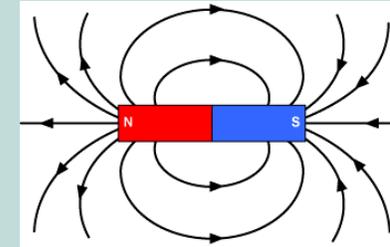


<https://bda668.wordpress.com/>

Temperature



Magnetic field



Pump-probe

Stroboscopic measurements

Look at repetitive processes



Current



Pressure/ strain

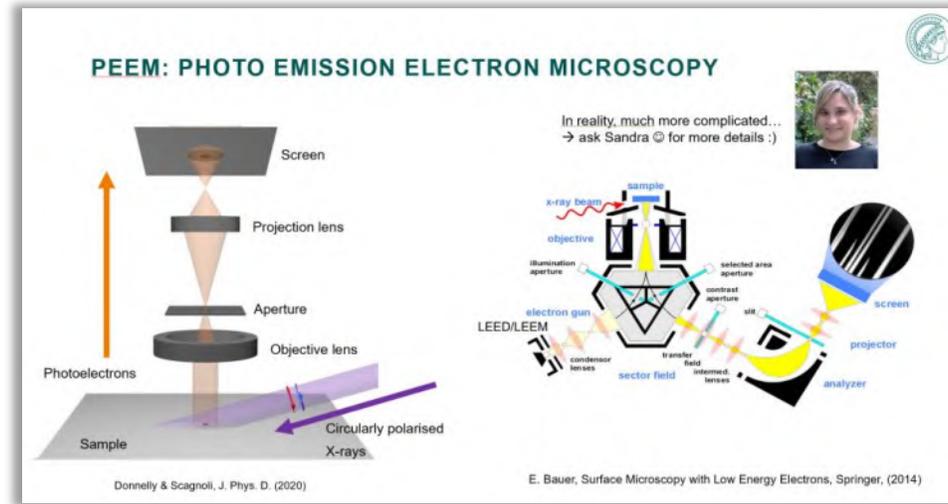




SINGLE SHOT: SUPERPARAMAGNETISM – OF NANOSTRUCTURES

Single-shot
Measurement sufficiently short to measure relevant dynamics
Need enough statistics!
Need to measure fast enough

<https://bda668.wordpress.com/>

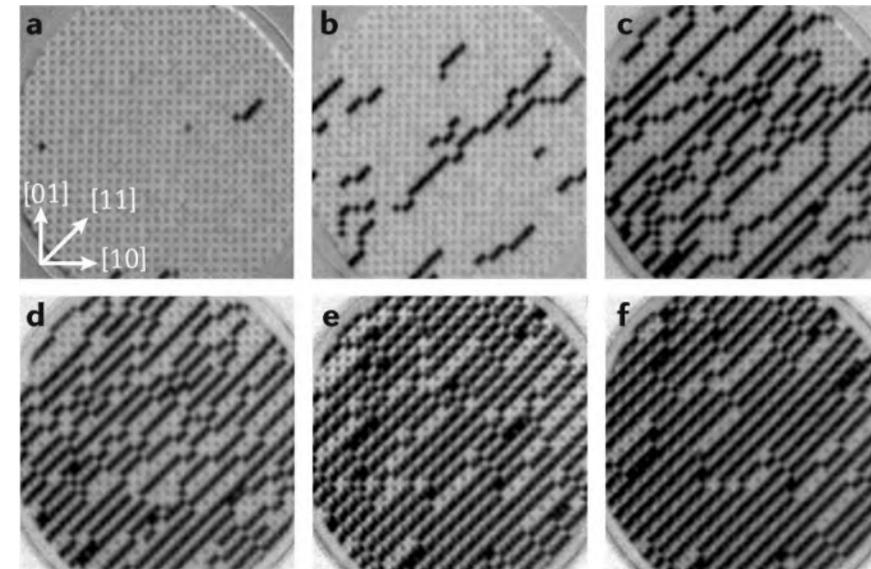
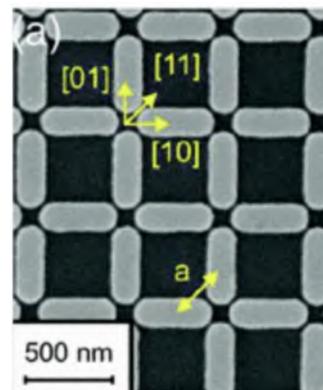


PEEM Measurement:
seconds → minutes

For patterned nanostructures

Blocking temperature: temperature at which the structures become thermally active.

Artificial spin ice: thermal evolution (slow!)



Farhan et al. Phys. Rev. Lett. 111, 057204 (2013).



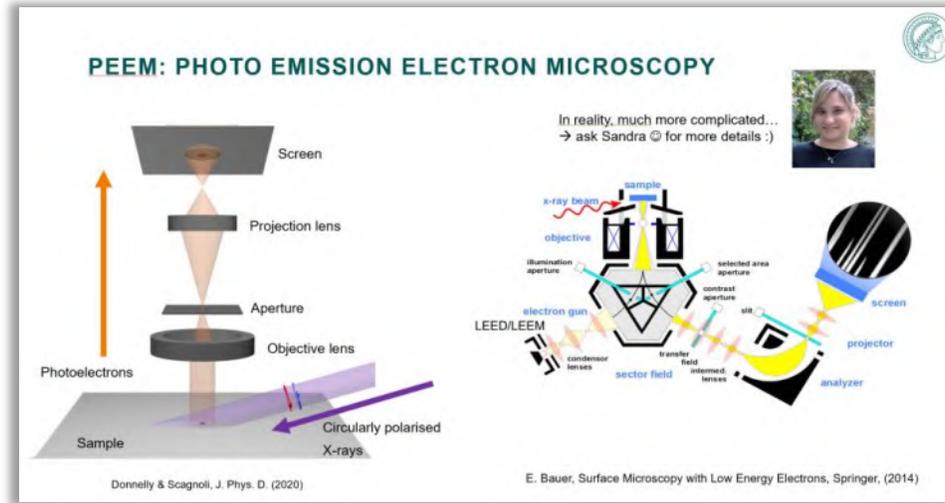
SINGLE SHOT/ QUASI-STATIC: NEED STATISTICS! SUPERPARAMAGNETISM – OF NANOSTRUCTURES

Single-shot

Measurement sufficiently short to
measure relevant dynamics

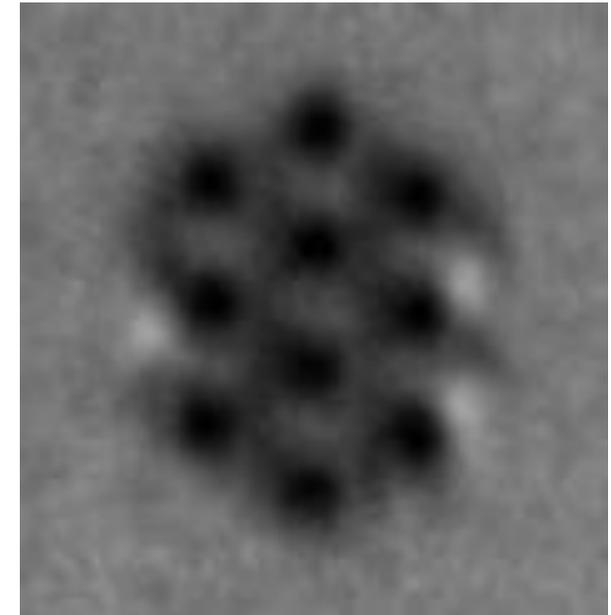
Need enough statistics!

Need to measure fast enough



PEEM Measurement:

seconds → minutes



<https://mesosys.mat.ethz.ch/research/artificial-spin-ice/asi-imaging.html>

For patterned nanostructures

Blocking temperature: temperature at which
the structures become thermally active.

Artificial spin ice: thermal evolution (slow!)



SINGLE SHOT: NEED STATISTICS! FASTER? → FREE ELECTRON LASER

Free electron laser

Very long accelerator: European XFEL 3.4 km long!



Ultra-bright, ultra-short X-ray pulses

10^{12} photons/s

~fs pulse length



FS RESOLUTION FOR DYNAMIC MEASUREMENTS?

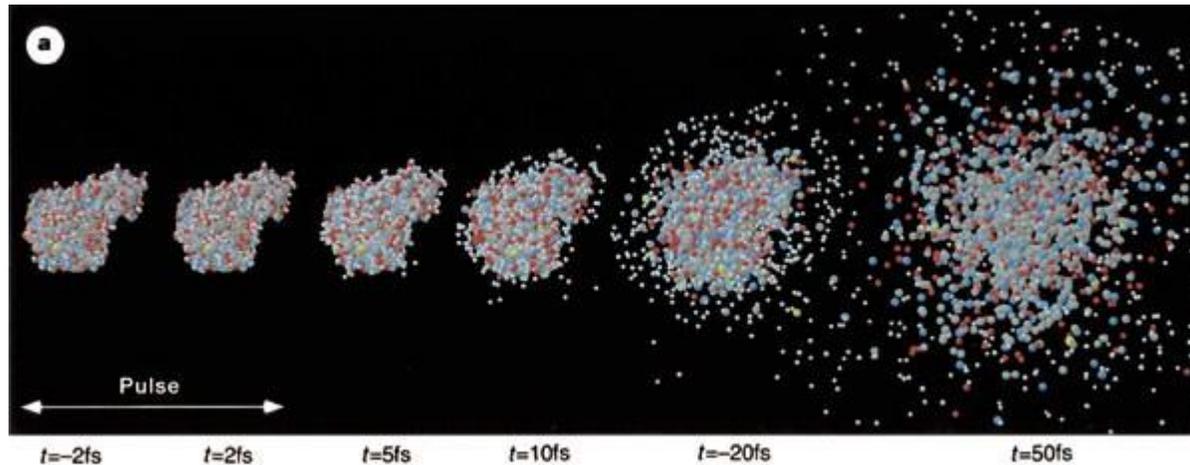
Ultra-bright, ultra-short X-ray pulses

10^{12} photons/s

~fs pulse length

Why? → damage/ non-reproducible dynamics

Powerful for many fields including biology



Neutze et al., Nature 406, 752 (2000)

Future: also apply to magnetisation dynamics, holography & CDI

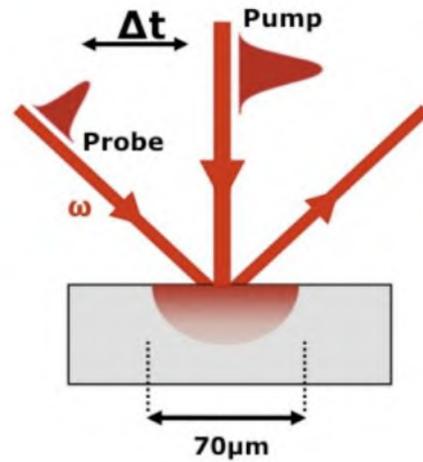
Challenge: fast detectors, dynamic range!



MORE STATISTICS: PUMP PROBE → STROBOSCOPIC

Pump-probe

Stroboscopic measurements
Look at repetitive processes

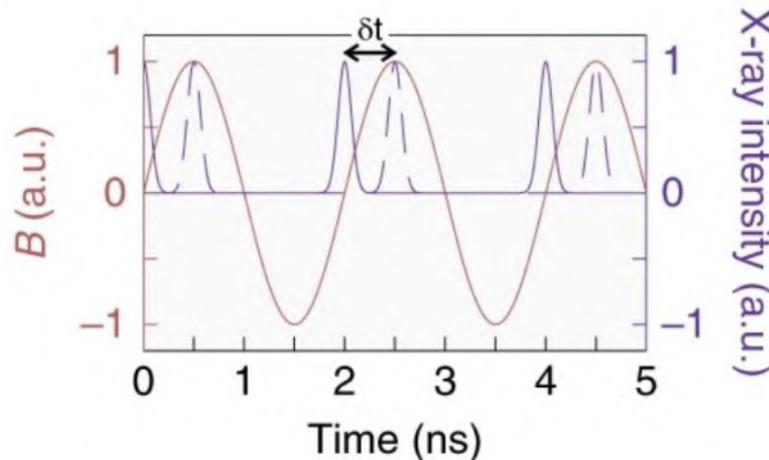


Key aspect:

Pump: excite the system

Probe: Measure the response of the system

Introduce a time delay in between pump & probe to map out dynamic response



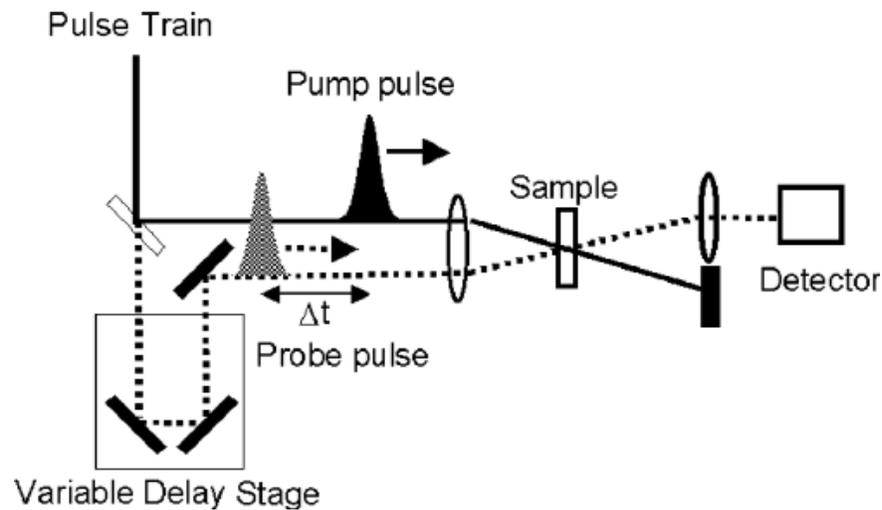
Temporal resolution of probes:

Laser	Synchrotron	XFEL
fs	ps	fs

DIFFERENT TYPES OF PUMP PROBE

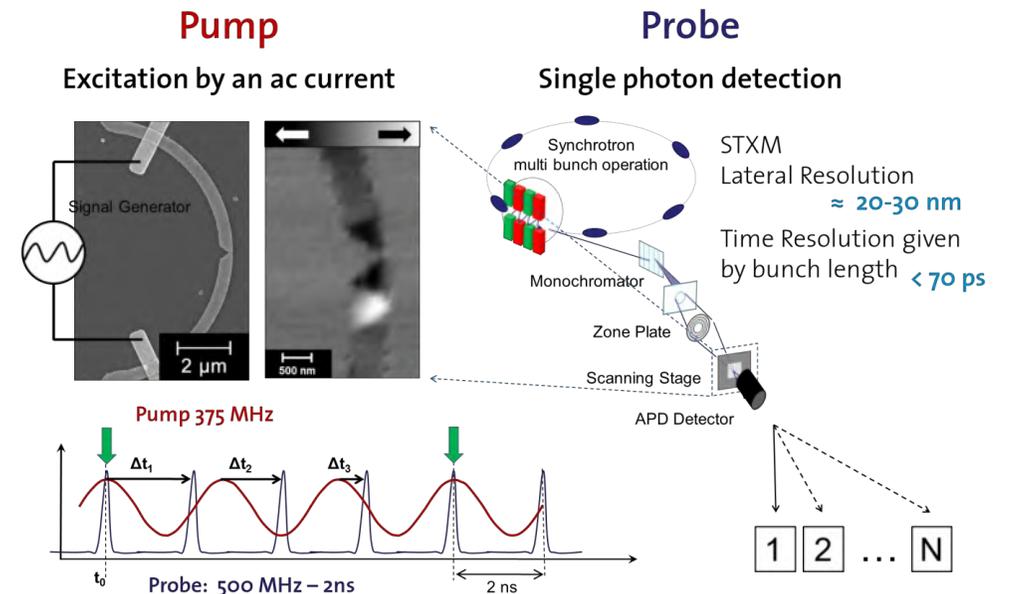
Single pump-single probe (classical method)

- Integrate signal for each time delay
- Require frequencies to match



→ Single pump-multiple probe (TR-STXM)

- Detect response to each individual excitation
- Pump and probe frequencies don't need to match!

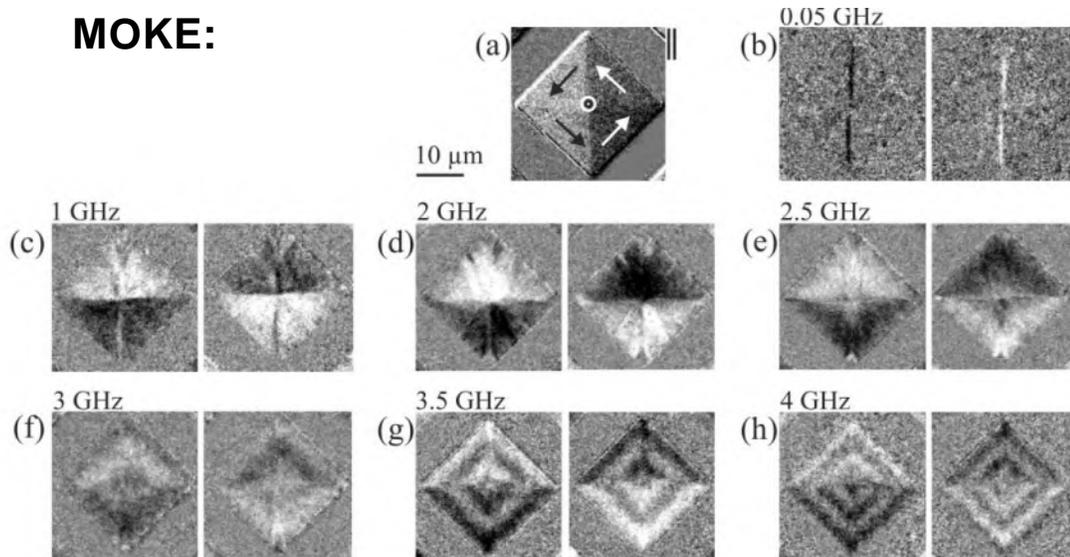


Images from: www.klaeui-lab.de

PUMP PROBE IMAGING OF SPIN WAVES

Apply pump-probe techniques to measure *coherent* spin waves:

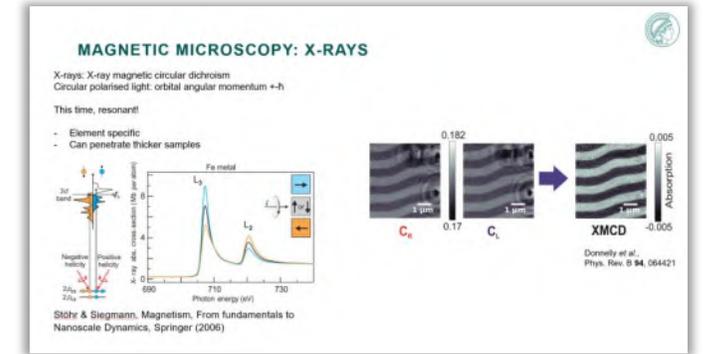
MOKE:



Time resolved MOKE

- Pump probe rotation of linear polarised light
- ps temporal resolution, 100s nm - μm spatial resolution

X-ray microscopy



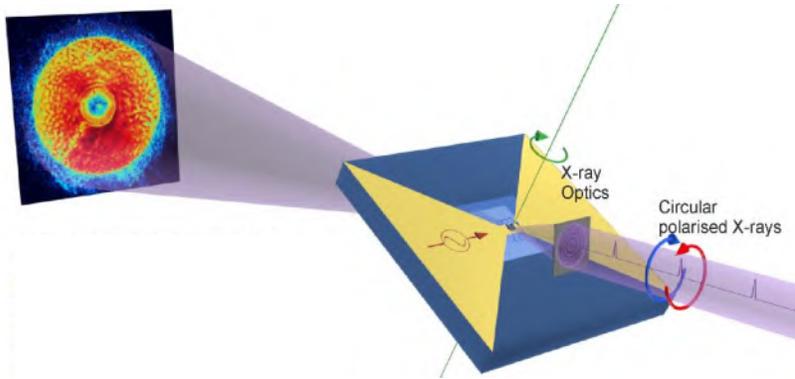
Synchrotron X-rays

Albisetti et al., *Sci Adv* 32, 1906439 (2020)

- Time resolved Scanning transmission X-ray microscopy
- Pump-probe XMCD
- Ps temporal resolution, nm spatial resolution

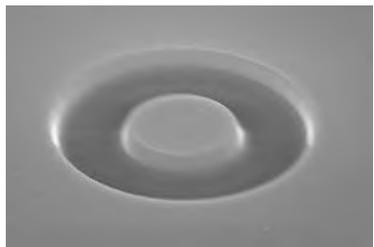
GOING TO THE 4TH DIMENSION:

X-ray magnetic laminography:

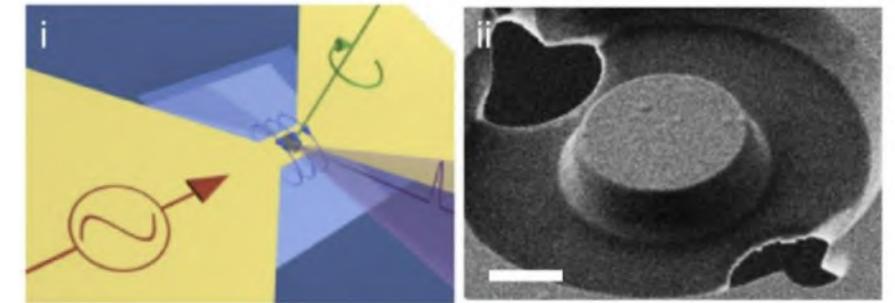
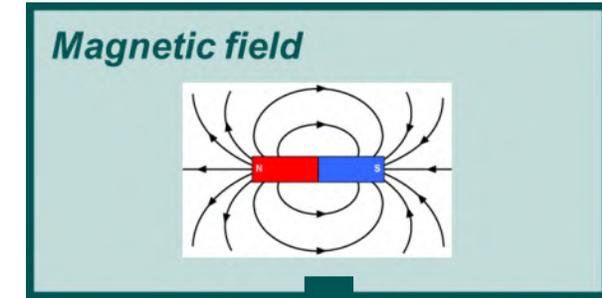


Combine 3D imaging and pump probe:

~100 ps temp. res.
~20 nm spatial res.



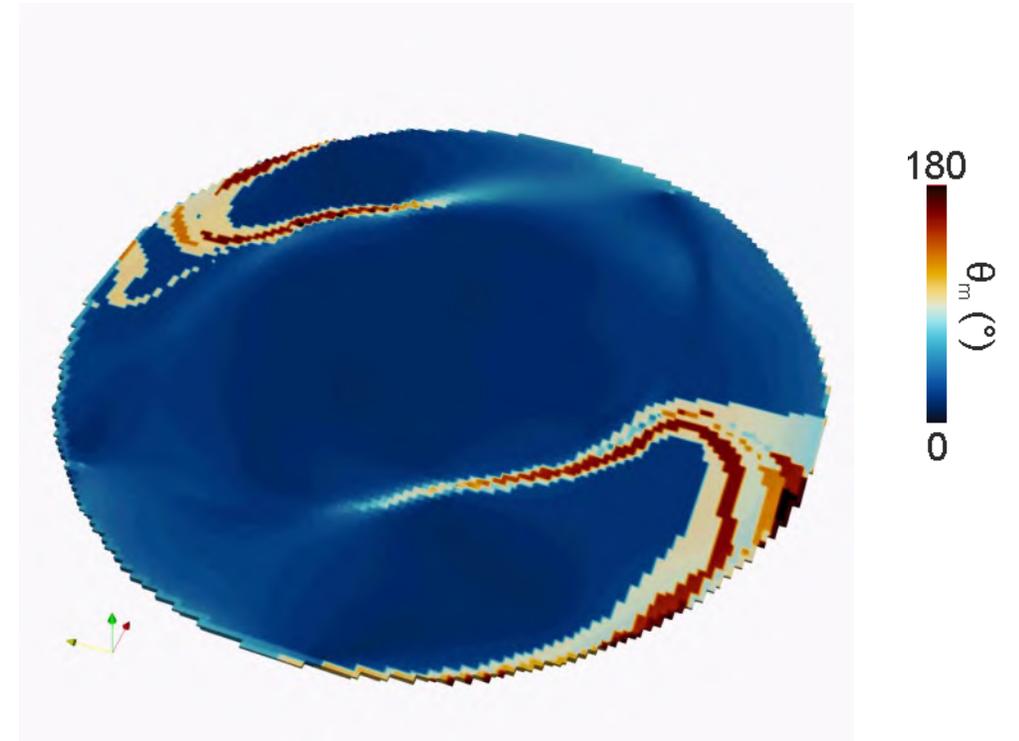
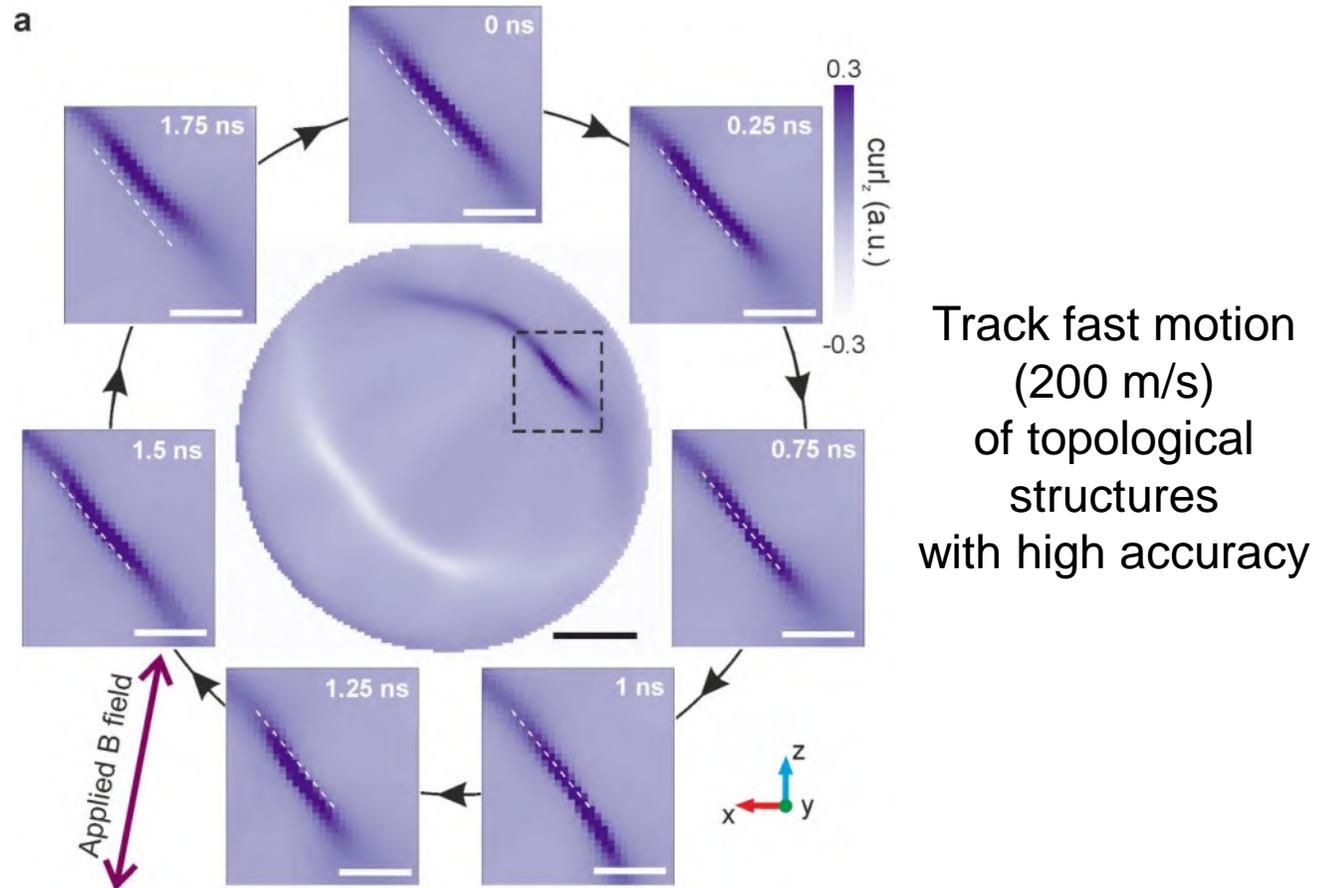
5 μm diameter disc
1.2 μm -thick GdCo



Oersted field of stripline

Donnelly et al., Nature Nanotechnology **15** 356 (2020)

MAPPING MAGNETISATION DYNAMICS IN 3D



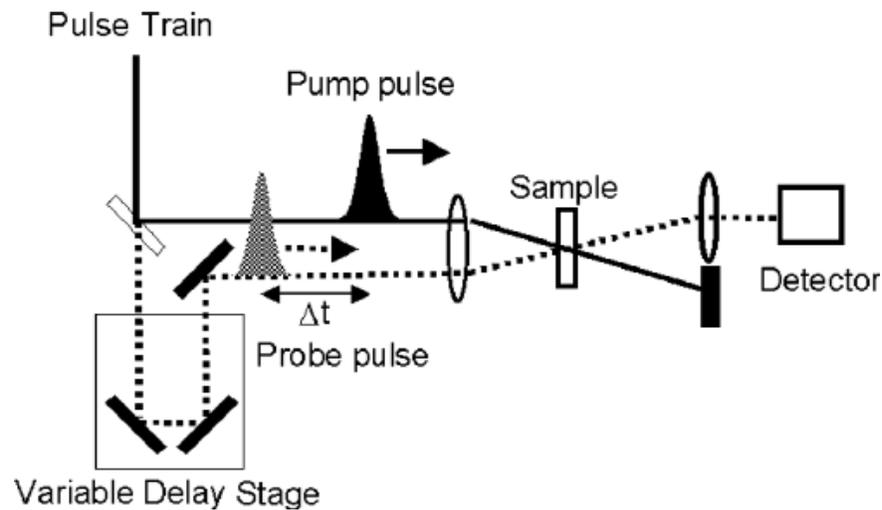
→ Map the modes of the magnetisation dynamics

Donnelly et al., Nature Nanotechnology **15** 356 (2020)

DIFFERENT TYPES OF PUMP PROBE

Single pump-single probe (classical method)

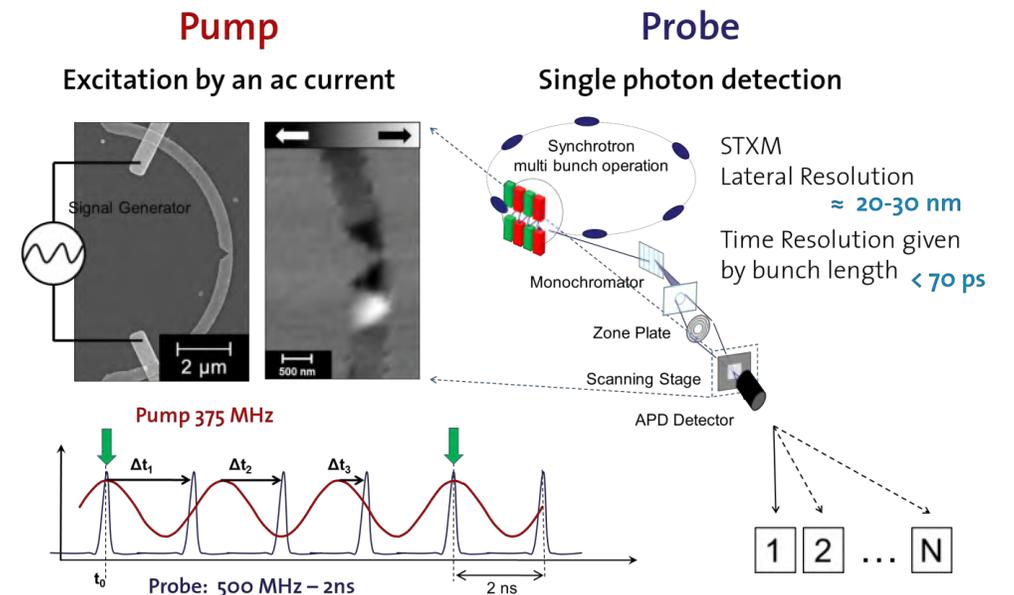
- Integrate signal for each time delay
- Require frequencies to match



Images from: www.klaeui-lab.de

→ Single pump-multiple probe (TR-STXM)

- Detect response to each individual excitation
- Pump and probe frequencies don't need to match!





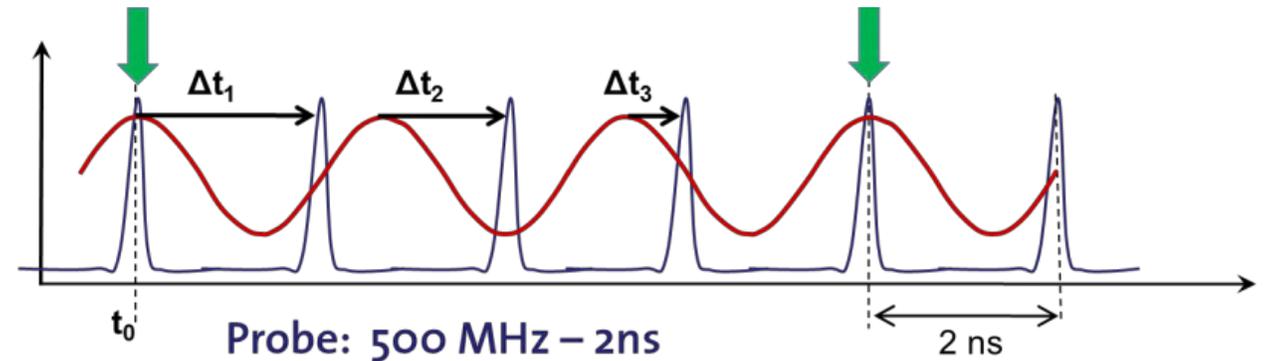
SINGLE PUMP MULTI PROBE: ARBITRARY FREQUENCIES

→ Single pump-multiple probe (TR-STXM)

- Detect response to each individual excitation
- Pump and probe frequencies don't need to match!

Very fast detector: Avalanche Photo Diode

- Photon bunches of the multibunch filling pattern (500 MHz repetition rate) can be resolved



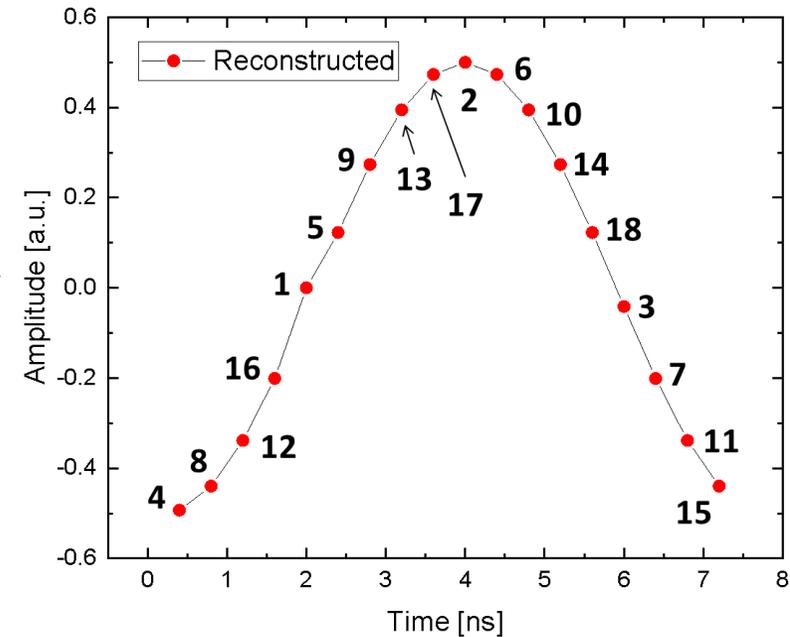
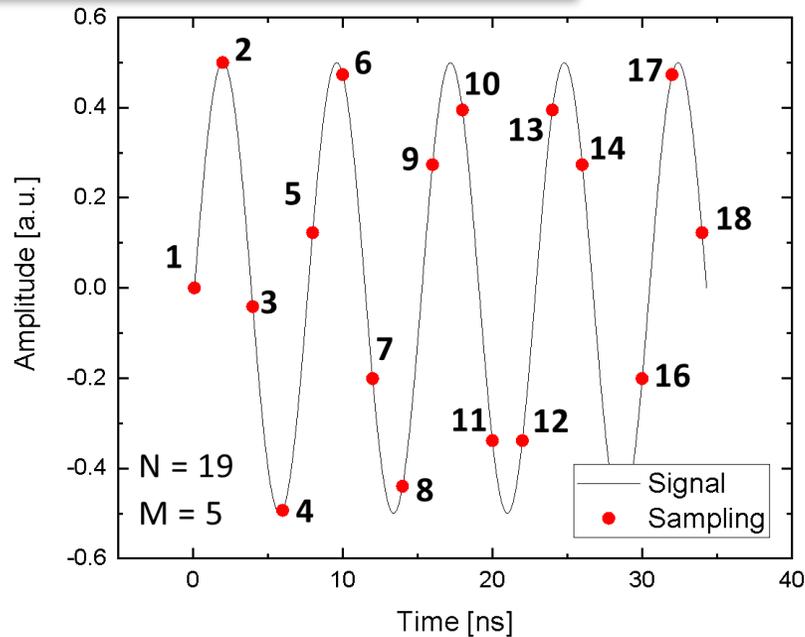
Images provided by Simone Finizio, PSI



SINGLE PUMP MULTI PROBE: ARBITRARY FREQUENCIES

→ Single pump-multiple probe (TR-STXM)

- Detect response to each individual excitation
- Pump and probe frequencies don't need to match!



For arbitrary frequency of excitation:

→ A temporal sorting of the acquired data is performed with fast electronics

Images provided by Simone Finizio, PSI



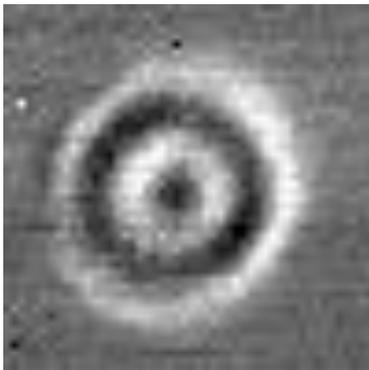
SINGLE PUMP MULTI PROBE: ARBITRARY FREQUENCIES

→ **Single pump-multiple probe (TR-STXM)**

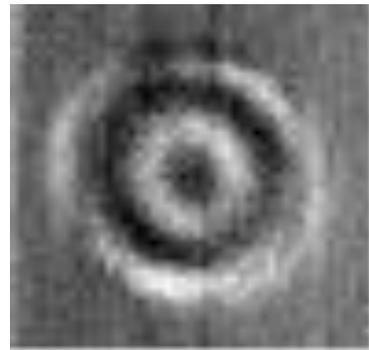
- Detect response to each individual excitation
- Pump and probe frequencies don't need to match!

Enables exploration of different dynamic modes in a sample!

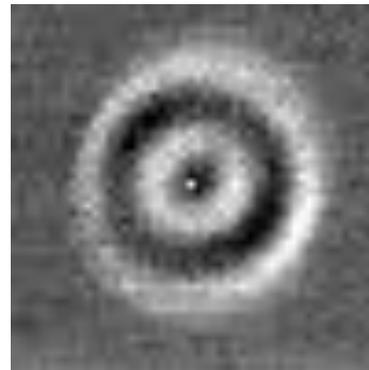
→ Here, a target skyrmion in a permalloy disc:



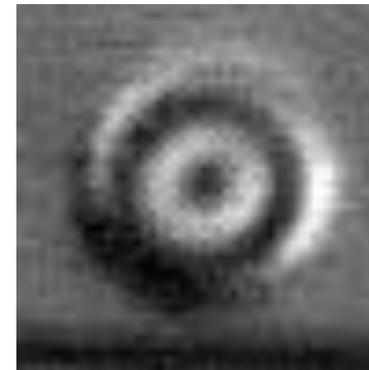
142 MHz
Gyration



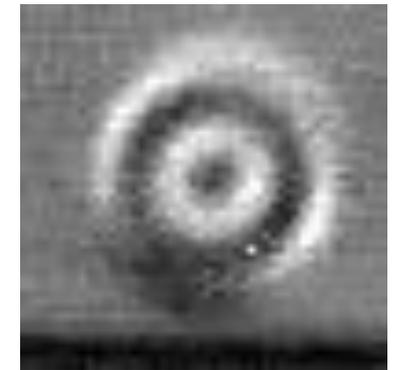
285 MHz
Gyration



785 MHz
Breathing



928 MHz
Gyration



1420 MHz
Gyration

1 μm

Images provided by Simone Finizio, PSI
Finizio et al., PRB 98, 104415 (2018)



SINGLE PUMP MULTI PROBE: ARBITRARY FREQUENCIES

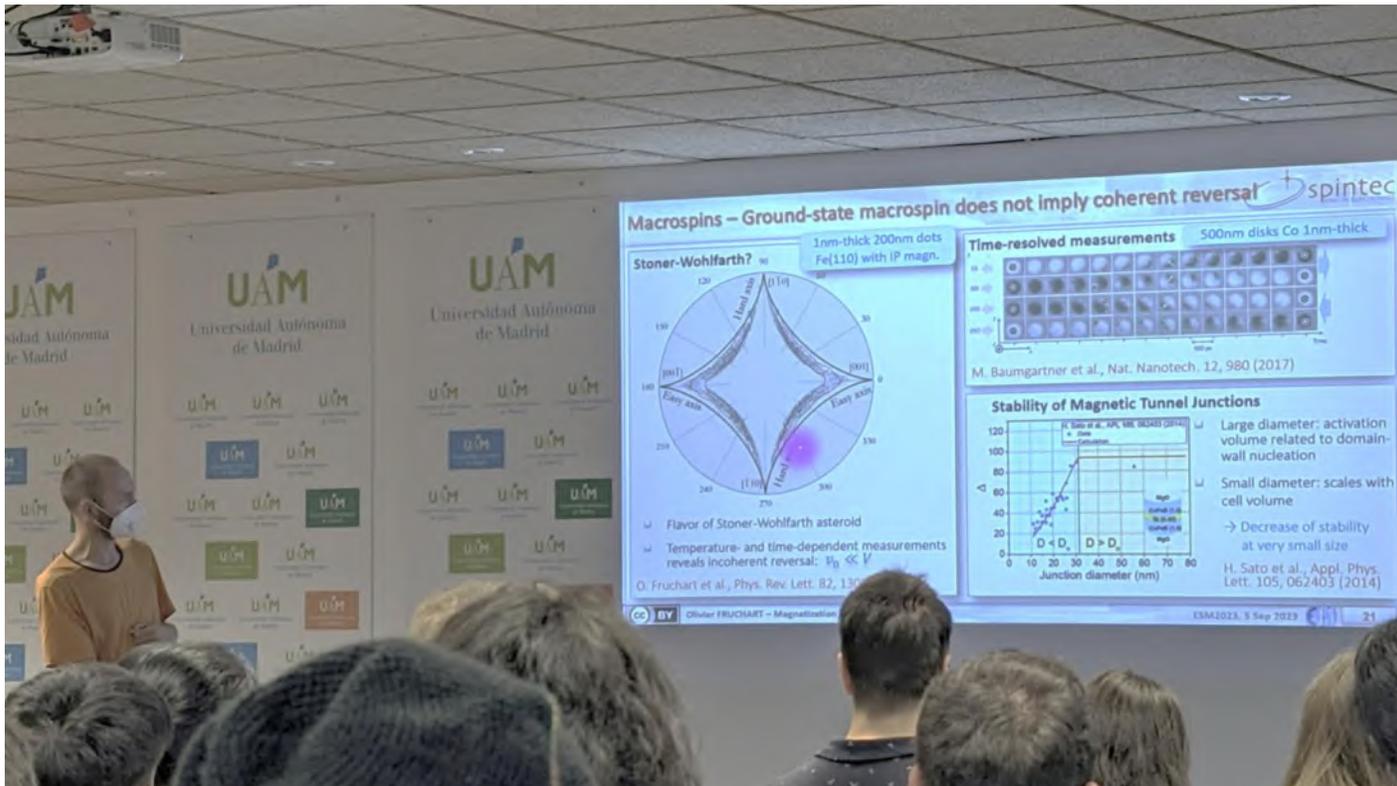
→ Single pump-multiple probe (TR-STXM)

- Detect response to each individual excitation
- Pump and probe frequencies don't need to match!

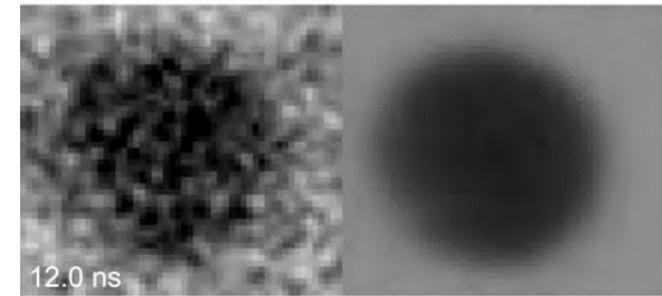
Enables exploration of different dynamic modes in a sample!

→ As well as the switching behaviour of systems:

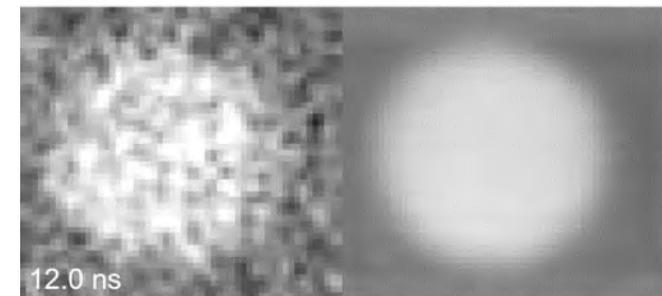
→ 1 nm thick Co discs:



SOT-induced magnetization switching



12.0 ns
100 nm $B_x = -124 \text{ mT}$, $\tau_p = 2 \text{ ns}$, $U_p = \pm 4 \text{ V}$



12.0 ns
100 nm $B_x = 94 \text{ mT}$, $\tau_p = 2 \text{ ns}$, $U_p = \pm 4 \text{ V}$

Baumgartner et al., Nat. Nano. 12, 980 (2017)

SINGLE PUMP MULTI PROBE: ARBITRARY FREQUENCIES

→ Single pump-multiple probe (TR-STXM)

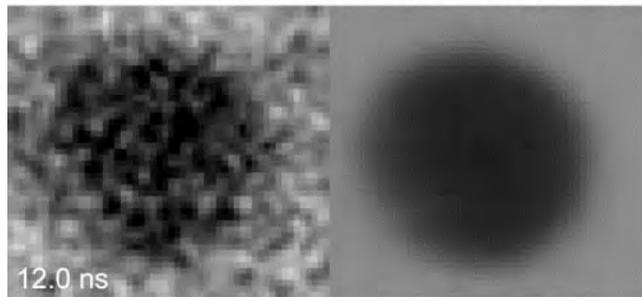
- Detect response to each individual excitation
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Enables exploration of different dynamic modes in a sample!

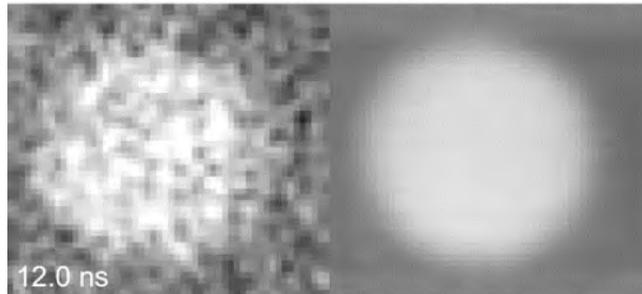
→ As well as the switching behaviour of systems:

→ 1 nm thick Co discs:

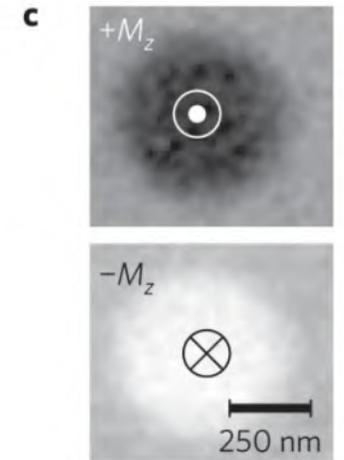
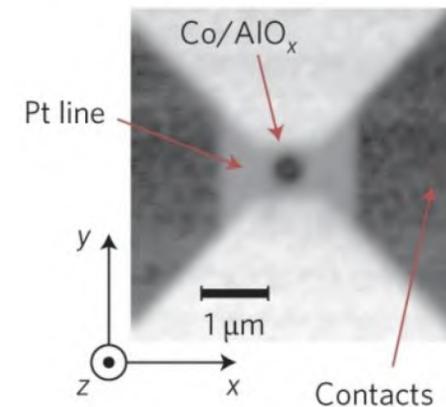
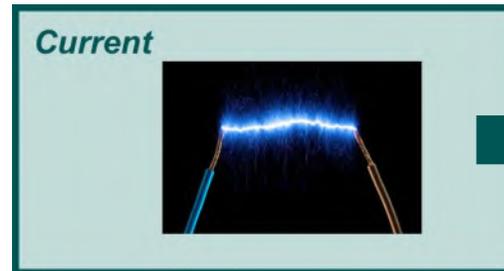
SOT-induced magnetization switching



12.0 ns
100 nm $B_x = -124 \text{ mT}$, $\tau_p = 2 \text{ ns}$, $U_p = \pm 4 \text{ V}$



12.0 ns
100 nm $B_x = 94 \text{ mT}$, $\tau_p = 2 \text{ ns}$, $U_p = \pm 4 \text{ V}$

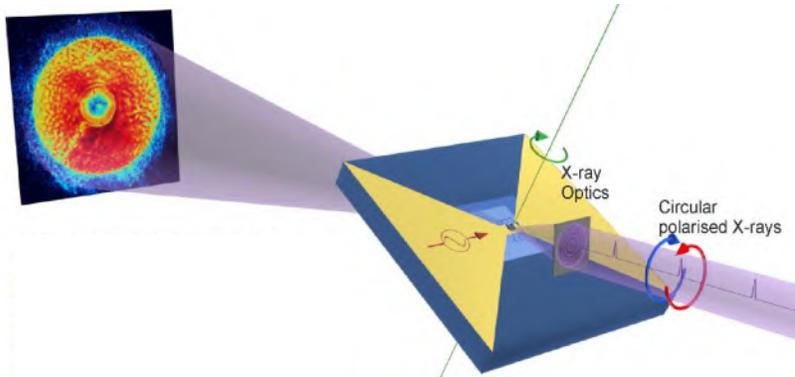


Baumgartner et al., Nat. Nano. 12, 980 (2017)



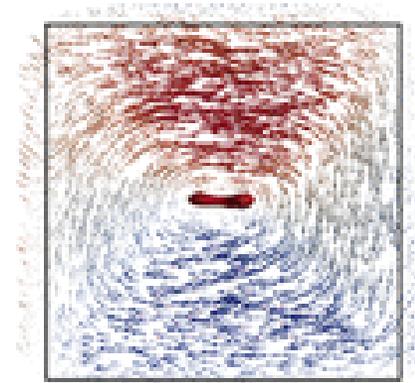
GOING TO THE 4TH DIMENSION:

X-ray magnetic laminography:

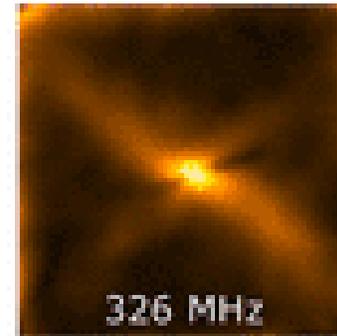


Combine 3D imaging and pump probe:

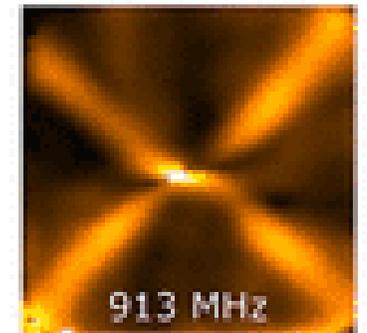
- ~ 100 ps temp. res.
- ~ 20 nm spatial res.



**Harness TR-STXM:
Image different resonant modes in 3D:**



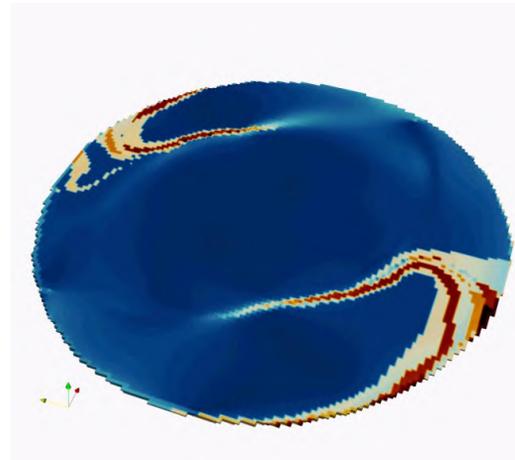
3D vortex core gyration



Domain wall excitation



Track topological structures with high accuracy



Map out coherent rotation modes

Donnelly et al., Nature Nanotechnology **15** 356 (2020)
Finizio, CD et al., Nano Letters **25**, 1971 (2022)

SPIN WAVES IN 3D?



Davide Girardi



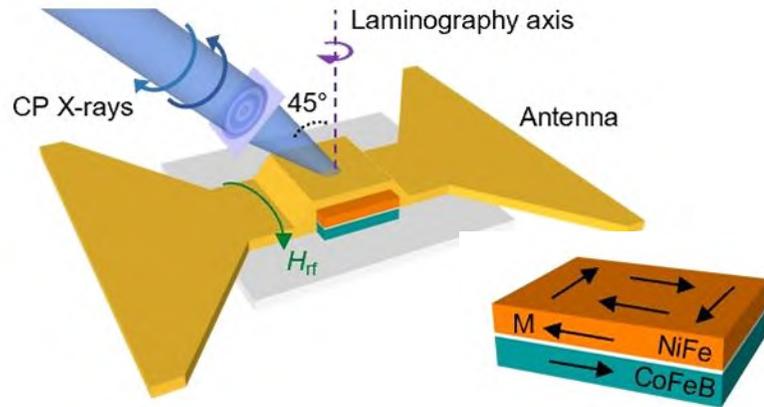
Edoardo Albisetti



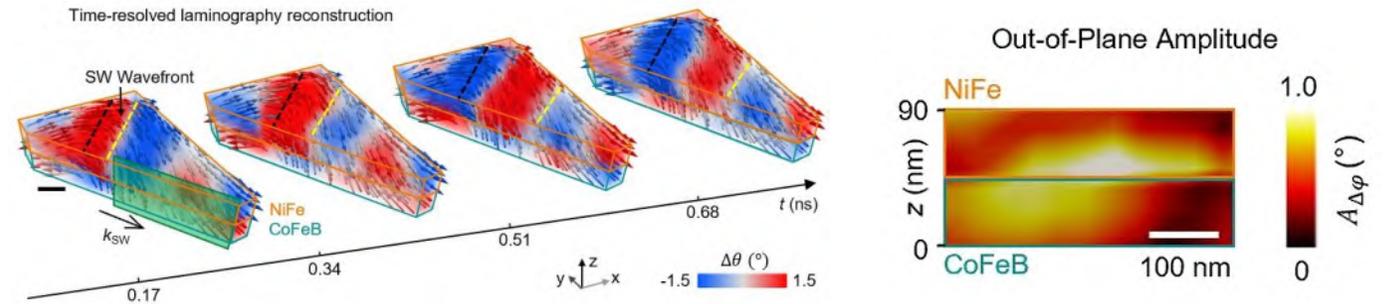
Daniela Petti



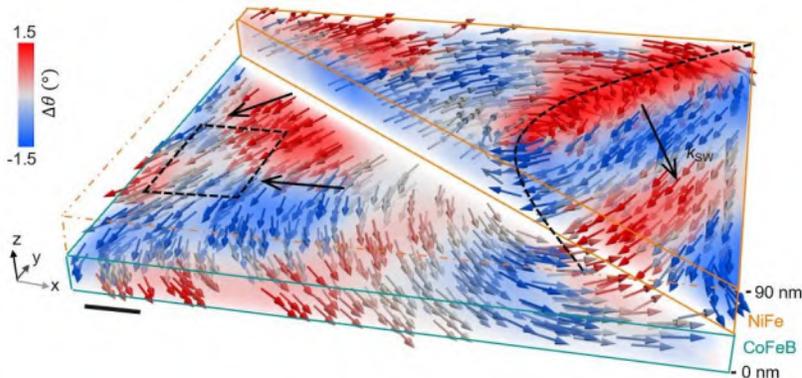
Resolving the three dimensional structure of spin waves in a SAF



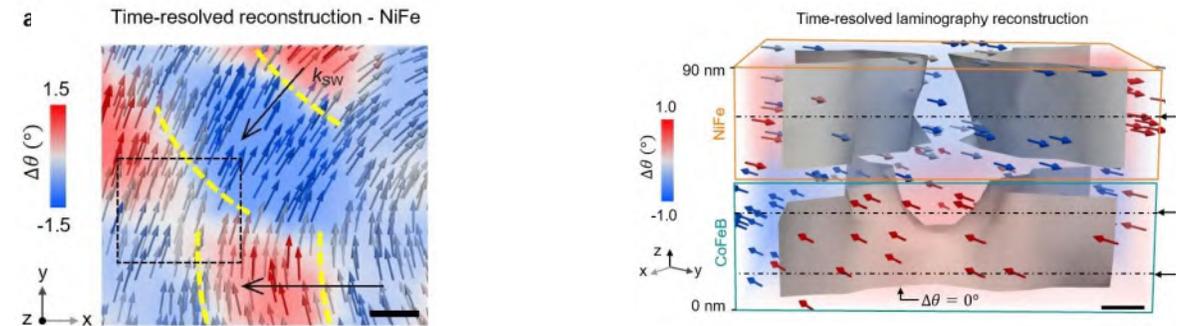
Localisation of propagating spin wave modes in 3D



Domain wall emits spin waves



Reveal 3D interference pattern of spin waves

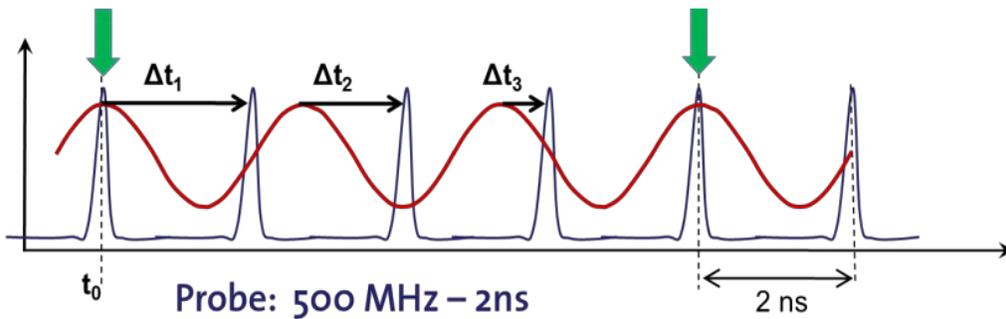


Girardi, CD et al., In review



TEMPORAL RESOLUTION OF SYNCHROTRON MEASUREMENTS: “TIME OF ARRIVAL STXM”

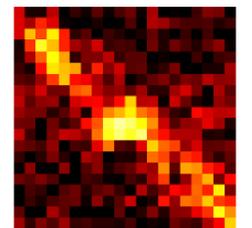
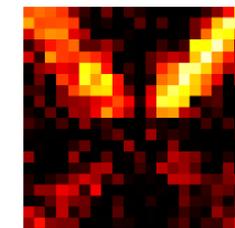
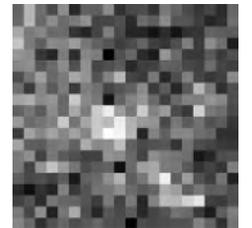
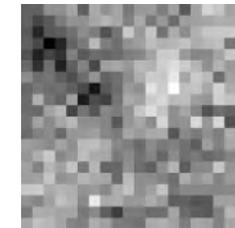
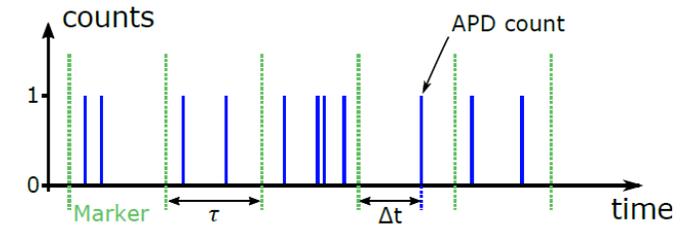
Electron bunches:
Finite width ~ 70 ps (SLS)



S. Finizio *et al.*, J. Syn. Rad. **27**, 1320 (2020)

What if we could detect each photon?

- Detect with 10 ps resolution
- 30 ps temporal resolution confirmed
 - Beyond the resolution of the synchrotron!
- Power of advanced electronics



500 nm

0 1
Normalized amplitude



OUR QUESTIONS FOR TODAY:

Higher dimensional investigations?

Vectorial imaging



Magnetisation dynamics

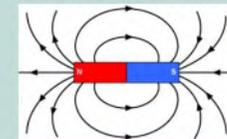


Advanced sample environments

Temperature



Magnetic field



Current



Pressure/ strain





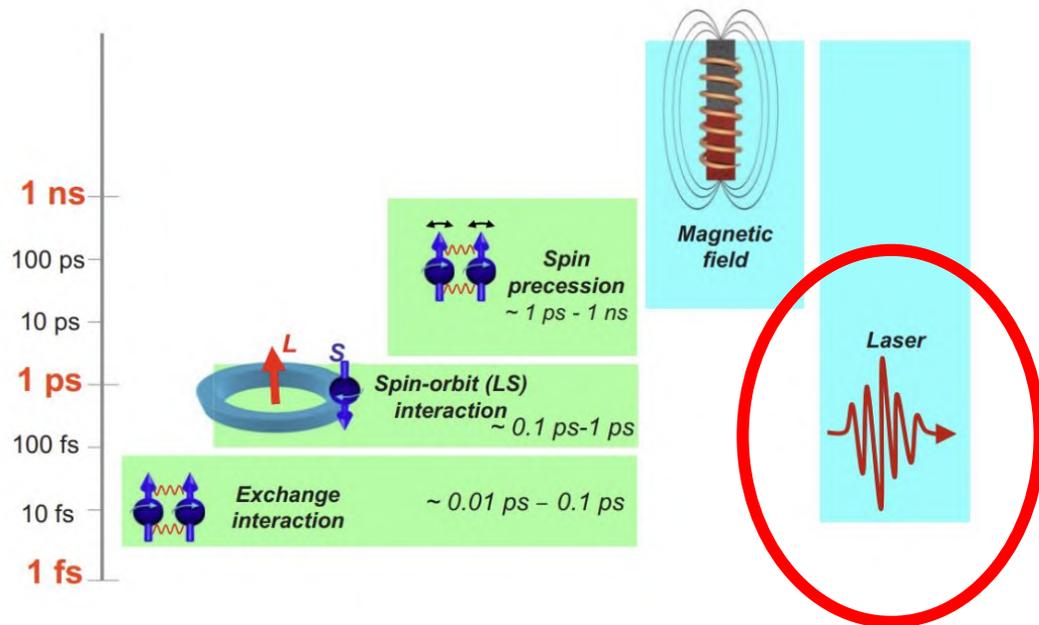
WHAT TIMESCALES ARE WE ON?

REVIEWS OF MODERN PHYSICS, VOLUME 82, JULY-SEPTEMBER 2010

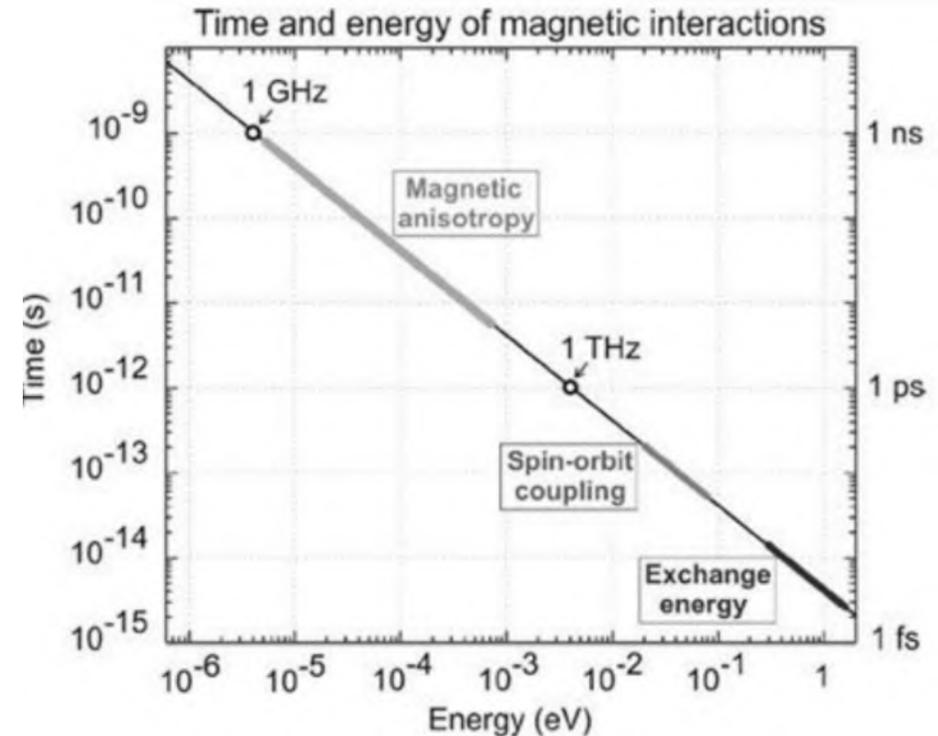
Ultrafast optical manipulation of magnetic order

Andrei Kirilyuk,^{*} Alexey V. Kimel, and Theo Rasing

Radboud University Nijmegen, Institute for Molecules and Materials, Heyendaalseweg 135, 6525 AJ Nijmegen, The Netherlands



There are faster time-scales out there!



Stoehr et al., *Magnetism* Springer-Verlag, (2006)

GOING ULTRAFAST



So far, we have looked at **thermal** and **magnetic field/ current** driven dynamics down to ps

→ However, when excited by a femtosecond laser, spin dynamics can be sub-pico second!

VOLUME 76, NUMBER 22

PHYSICAL REVIEW LETTERS

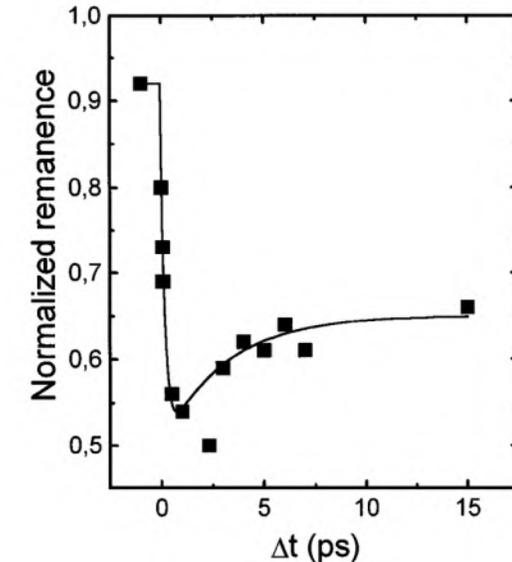
27 MAY 1996

Ultrafast Spin Dynamics in Ferromagnetic Nickel

E. Beaurepaire, J.-C. Merle, A. Daunois, and J.-Y. Bigot

*Institut de Physique et Chimie des Matériaux de Strasbourg, Unité Mixte 380046 CNRS-ULP-EHICS,
23, rue du Loess, 67037 Strasbourg Cedex, France*

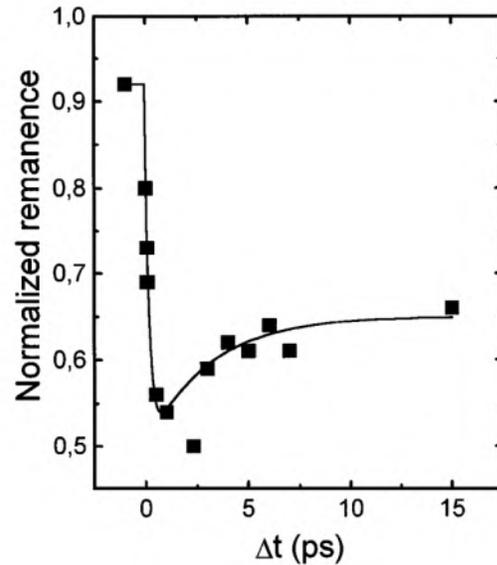
(Received 17 October 1995)



FEMTOSECOND DYNAMICS: ULTRAFAST DEMAGNETISATION



Nickel

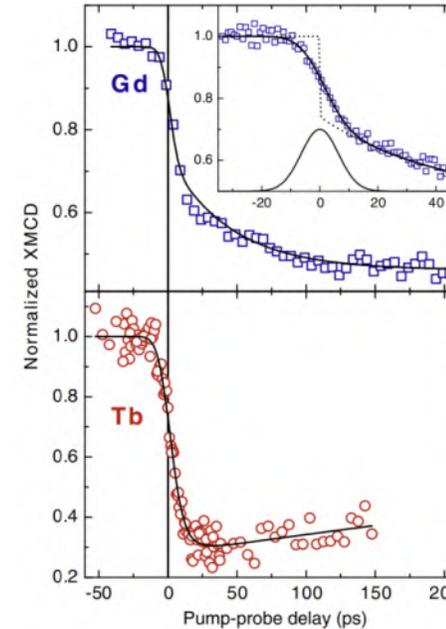


→ Demagnetisation in sub-ps time scale

→ Recovers ~ ps

Beaurepaire et al., Phys. Rev. Lett. 76, 4250 (1996)

Gadolinium



→ Demagnetisation in 10/100 ps timescale

Wietstruk et al. Phys. Rev. Lett. 106, 127401 (2011)



ULTRAFAST: WHAT'S HAPPENING?

Longer time scales:

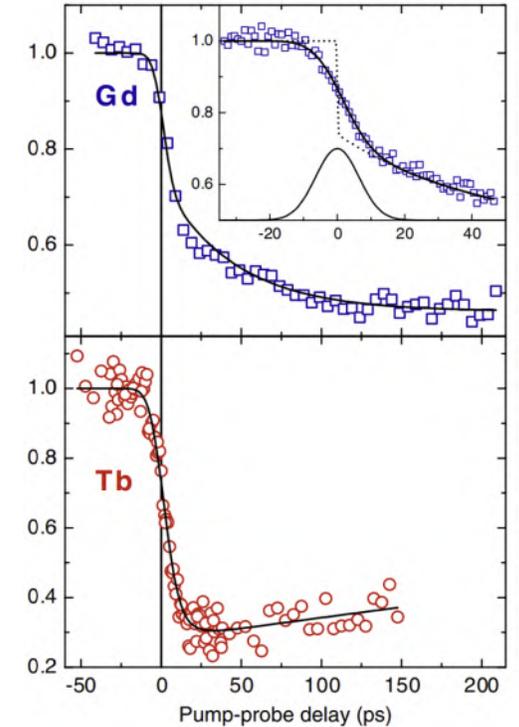
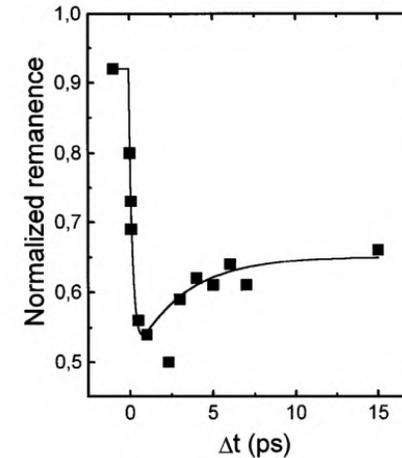
Spin & lattice in equilibrium

Fast time scales:

Lattice-driven spin demagnetisation!

Electrons act as heat bath for spin system

→ **Effective heating of the spins above the Curie temperature**



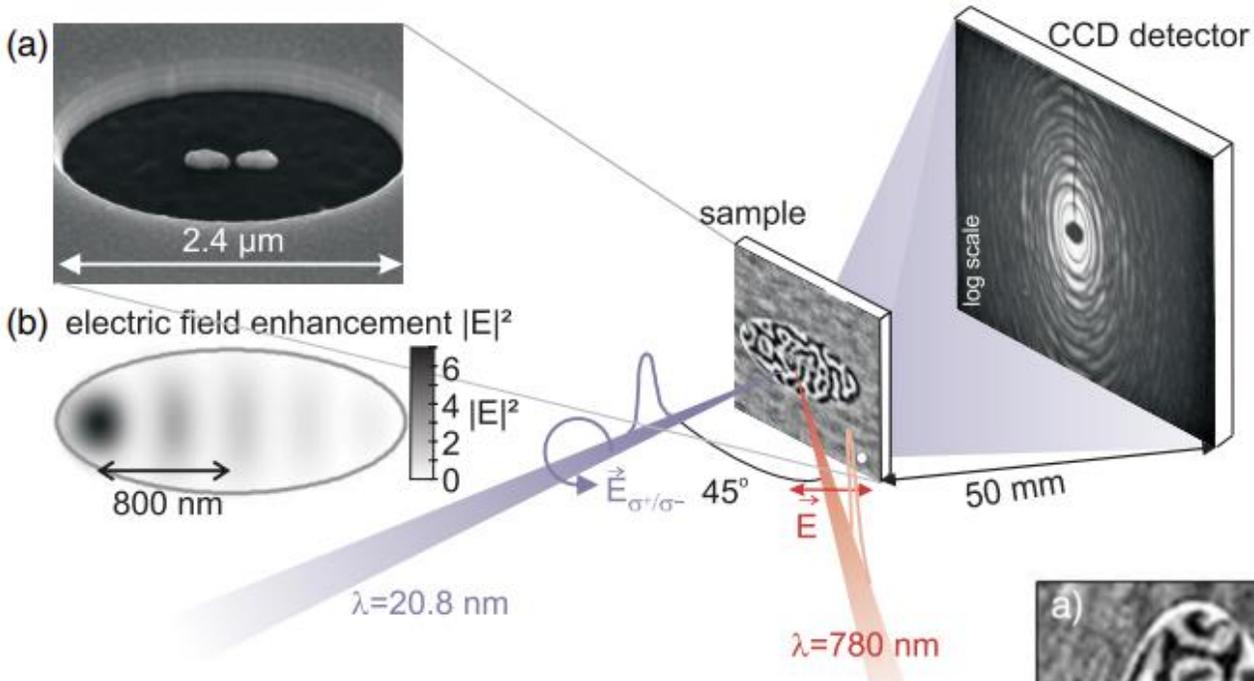
Ni vs Gd?

Magnetic moment ~ Angular momentum

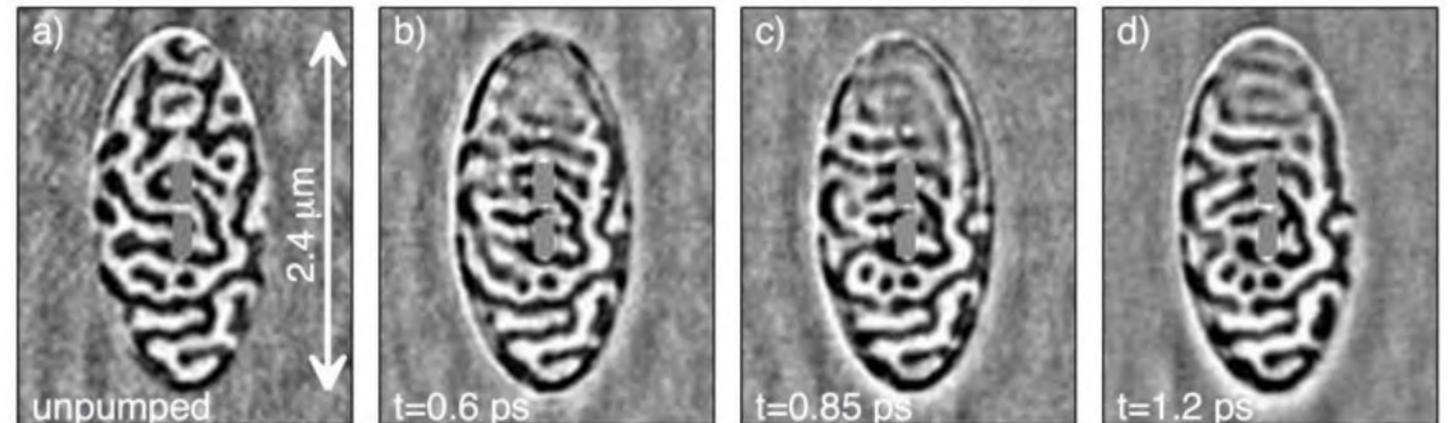
Moment of Gd larger (7.3 vs 1.26 μB)

Rate of change of L ~ similar

ULTRAFAST: WHAT'S HAPPENING? → X-RAY HOLOGRAPHY

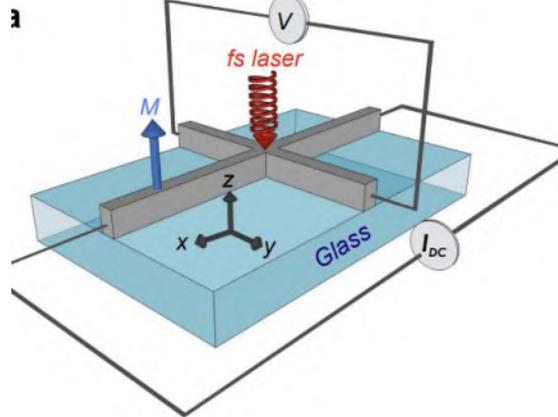


**Resolve demagnetization propagation front:
Moves at 0.2 nm/fs**

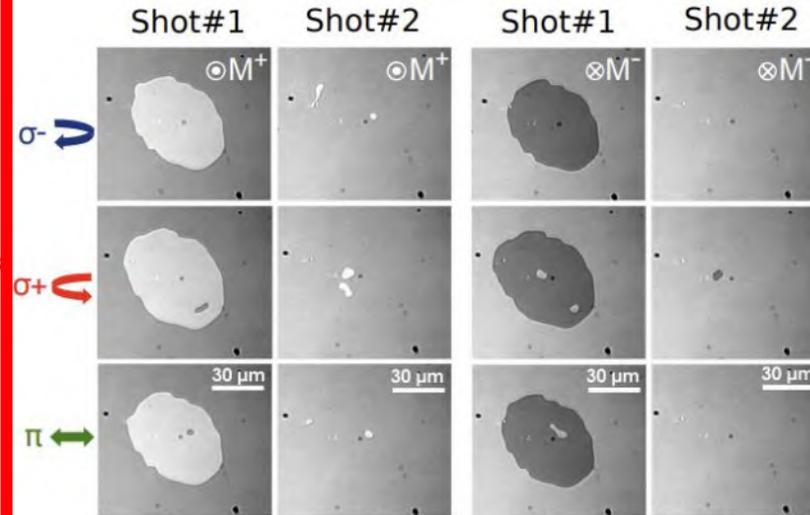


C. von Korff Schmising PRL 112, 217203 (2014)

ULTRAFAST DYNAMICS: SWITCHING?



Ferrimagnetic alloy, GdFeCo

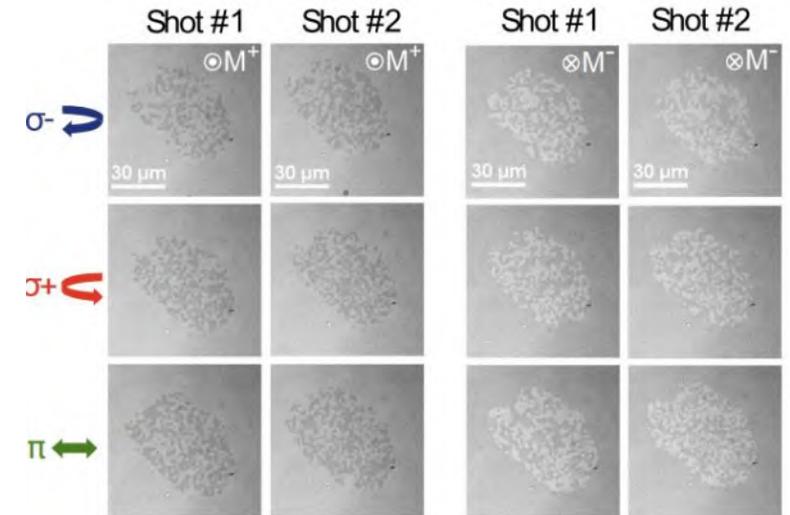


→ Helicity independent, single shot switching

→ Single shot: how fast?

Fast switching!

Ferromagnetic Co/Pt multilayers



→ Multi-pulse, helicity dependent optical switching

→ ~ms

El Hadri et al, Phys. Rev. B , 94, 064412 (2016)

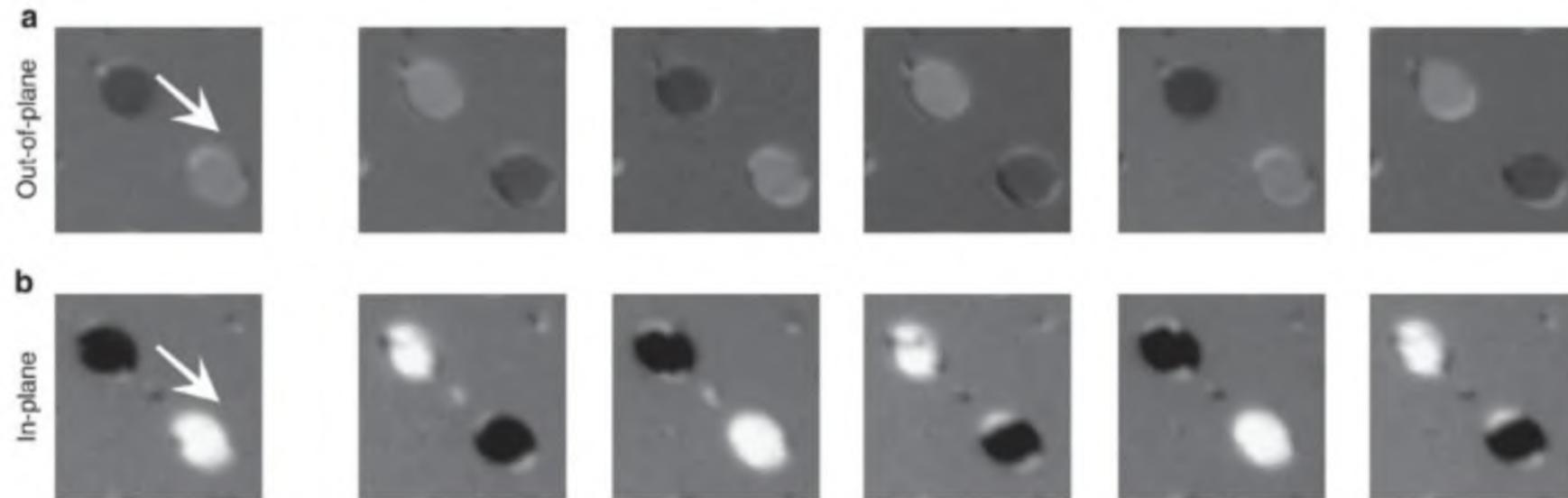


ALL-OPTICAL SWITCHING

GdFeCo:

Reproducible switching back and forth

Imaging with MOKE



Ostler et al., Nat. Commun. 3, 666 (2012)



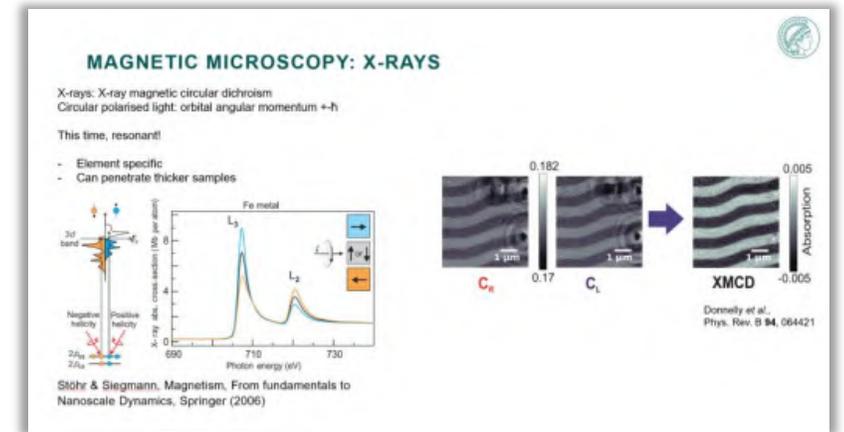
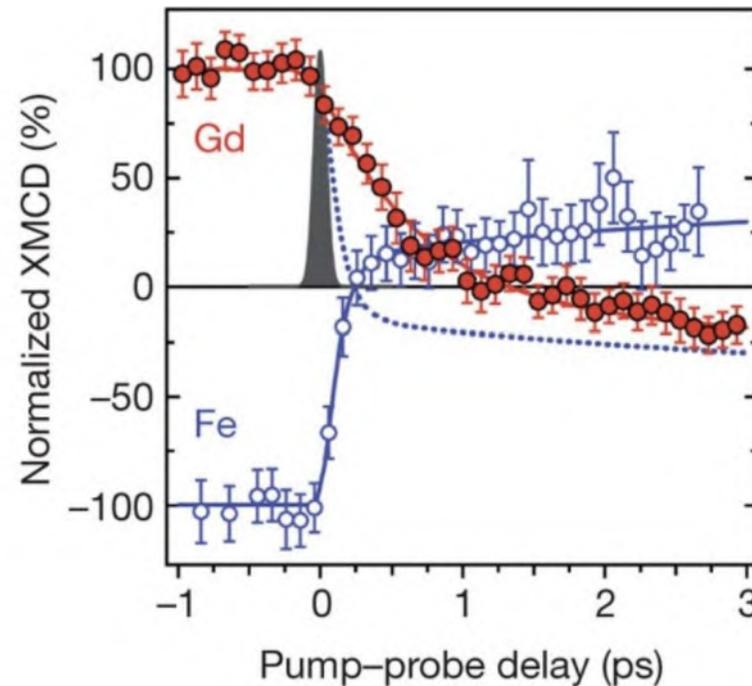
ULTRAFAST ALL-OPTICAL SWITCHING

Key: ferrimagnet?

Use element-specific XMCD probe

Femto-slicing beamline for soft x-rays

Reveals:



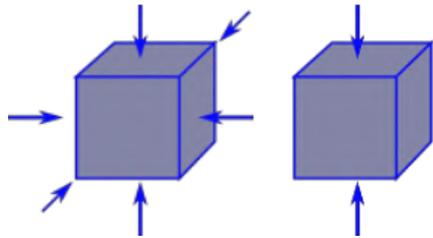
Fe and Gd demagnetise on different timescales
→ Transient ferromagnetic state
→ Drives robust switching process

Radu et al., Nature **472**, 205 (2011)

APPLICATION OF PRESSURE: FAST & SLOW



Physical pressure

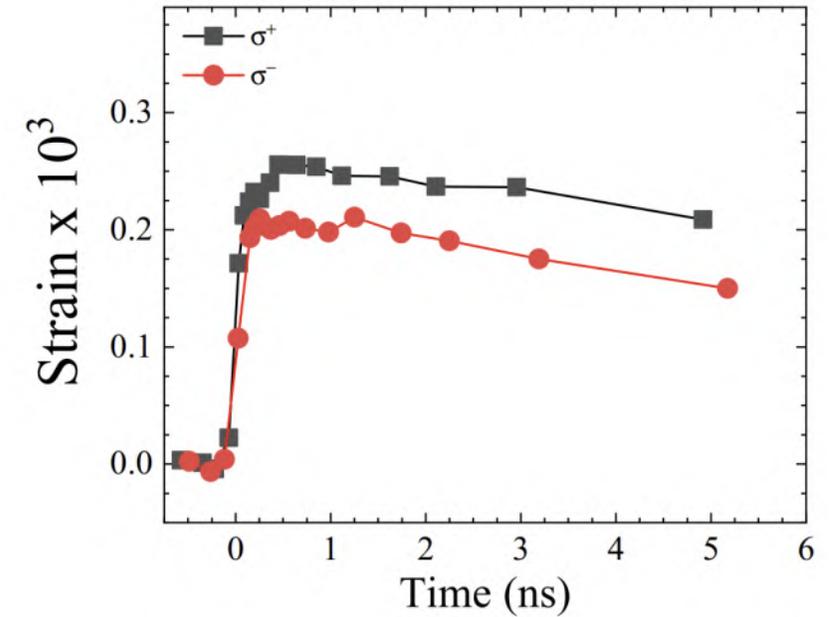


Diamond Anvil Cell



Possible to apply up to ... in situ
Mechanical motion → slow

Time-resolved lattice dynamics



Laser induced control of strain → fast!



OUR QUESTIONS FOR TODAY:

Higher dimensional investigations?

Vectorial imaging

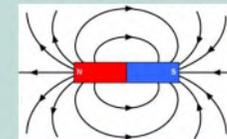
Magnetisation dynamics

Advanced sample environments

Temperature



Magnetic field



Current



Pressure/ strain





OUR QUESTIONS FOR THIS MORNING'S LECTURES:

What do we need to consider?

Spatial resolution

Sample environments

Time resolution

What methods are available?



Choosing the method for me and my samples?

